FINAL REPORT

Estimated Airborne Releases of Plutonium during the 1957 Fire in Building 71

Task 2: Verification and Analysis of Source Terms

August 1999

Submitted to the Colorado Department of Public Health and Environment, Disease Control and Environmental Epidemiology Division, Rocky Flats Health Studies in Partial Fulfillment of Contract No. 100APPRCODE 391

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SUMMARY

This investigation into the atmospheric releases during the 11–12 September 1957 fire in Building 71 (now called Building 771) is a part of Phase II of the Historical Public Exposures Studies on Rocky Flats. The study was performed by *Radiological Assessments Corporation* for the Colorado Department of Public Health and Environment. The release estimates are based upon information about the amount of plutonium involved in the fire, conditions before and during the fire, and release fractions for plutonium under various oxidation conditions. Estimates of the amount of plutonium released to the atmosphere are needed because stack effluent sampling capability was lost during the fire. No measurements of the amounts of airborne plutonium in gaseous effluents were made.

The fire, which began at about 10 p.m., was initiated by spontaneous combustion of impure plutonium in a metal casting residue (called a skull) stored in a glovebox line in Room 180. Portions of the gloveboxes were constructed of Plexiglas[a], which caught fire and burned as did rubber gloves attached to gloveboxes. Before the fire broke through the glovebox containment, heat and contaminated air were carried into the glovebox exhaust filter system. Small, combustible chemical warfare system (CWS) filters were installed in the glovebox outlet lines to remove plutonium from the glovebox exhausts. The fire burned through these filters quickly and spread to eight larger (2-feet square) combustible CWS filters that were part of the glovebox exhaust or booster system. The booster system filters also burned through and the fire then spread to the main filter bank, which contained more than 600 large CWS filters. Exhaust fans, located on the downstream side of the filters, discharged into a large duct that connected at the wall of the building with an underground duct that carried the contaminated exhaust air to the stack and to the environment.

The fire in Room 180 was discovered at 10:10 p.m. and was extinguished using water about 30 minutes later. The most affected work area was Room 180 and adjoining rooms, but all parts of the building were contaminated when an explosion or deflagration occurred in the ventilation system. This occurred shortly after the fire on the first floor was put out. A power failure terminated operation of the exhaust fans and the effluent sampler at about the same time. Firemen began to fight the fire in the main filter plenum at 11:15 p.m. That fire was declared "knocked down" at 2 a.m. Smoldering of filters and further application of water continued for several more hours. The fire was formally declared "out" at 11:28 a.m.

Figure S-1 is a schematic drawing that shows the atmospheric release pathways for plutonium during the fire. Plutonium was carried to the main filter plenum in two ways: (1) from the gloveboxes through the glovebox exhaust (booster) filtration system and (2) via the Room 180 ventilation system. Fire damage to the main plenum filters permitted large discharges to the environment via the stack.

Estimated releases from the 1957 fire and the associated uncertainties have been computed for 15-minute time intervals between 10 p.m. on 11 September and 2 a.m. on 12 September, when the plenum filter fire was declared to be knocked down. By 2 a.m., water had been applied to the plenum filter fire for more than 2 hours. By that time, releases from the plenum filter fire were small compared to those that occurred earlier. The sum of all releases after 2 a.m. was a small fraction of the total release.

[a] Plexiglas is a trade name of Rohm and Haas for several types of clear polymethylmethacrylate.

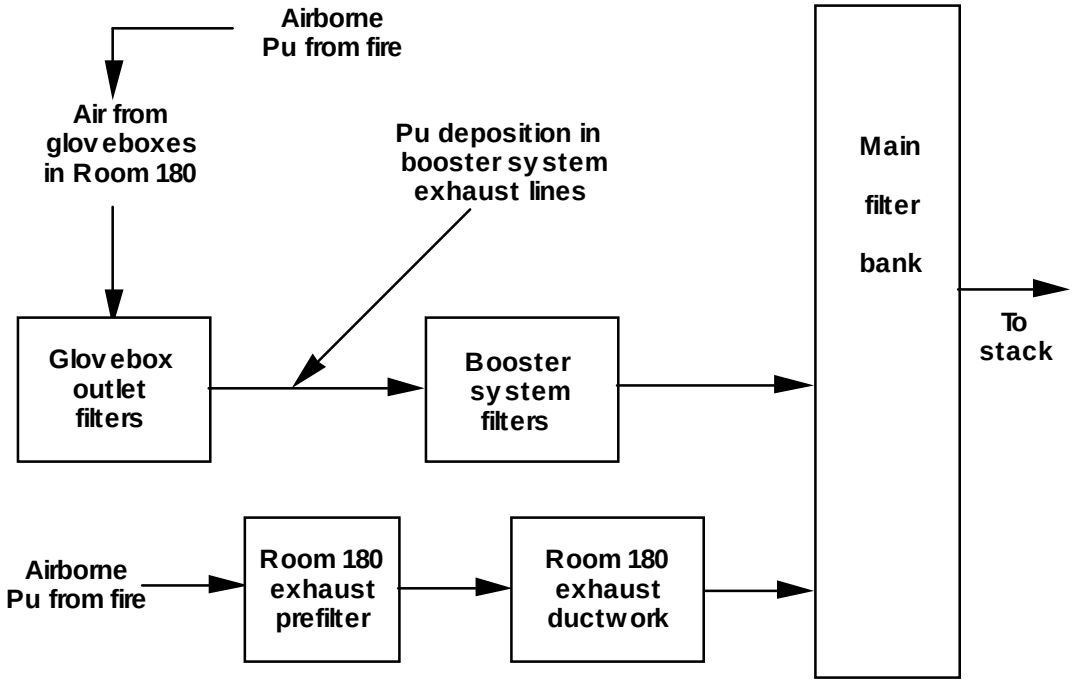

Figure S-1. Pathways for release of plutonium during the 1957 fire.

Analysis, based upon post-fire experimental evidence, indicates that the combustible CWS filters used in various effluent treatment systems burned rapidly. Data from tests, in which filters and Plexiglas were ignited and burned, were used to estimate the progress of the fire and to define times of filter failures. The largest releases occurred after the main filter bank was compromised. The period of highest release was estimated to be between 10:15 p.m. (before the primary fire was extinguished) and 10:45 p.m. (before water was applied to the plenum filter fire). This period is shown clearly in Figure S-2, which contains the estimated 15-minute average releases during the entire period from 10 p.m. to 2 a.m. The 15-minute interval was chosen to match the available meteorological data and, thereby, facilitate the calculations of environmental transport and risk. The central or median estimate (50^{th} percentile value) and the 5^{th} and 95^{th} percentiles of the distribution of estimates for each time period are all shown in the plot. After 10:45 p.m., the primary fire was out, the exhaust fans were off, and releases were much lower. After that time, the principal contribution to the releases was from the burning of the main plenum filters.

Figure S-3 shows the estimated releases during the first 45 minutes in more detail. As in Figure S-2, the median estimate and the 5^{th} and 95^{th} percentiles of the distribution of estimates are indicated. The time sequence of 1-minute average releases shown in this figure is uncertain but serves to illustrate the sequence of events. The period during which the plenum filters had caught fire and were burning through is highlighted. The estimated releases increase rapidly during that period. By the end of that time, the plenum filters were no longer effective in reducing the release from the fire in Room 180. The estimated releases continued at a high rate until the fire in Room 180 was extinguished (shortly before the exhaust fans stopped operating).

The explosion or deflagration also occurred just before the power to the fans was lost. The pressure pulse, which occurred after many plenum filters had already been severely damaged by fire, is not estimated to have had a significant effect on the plutonium release.

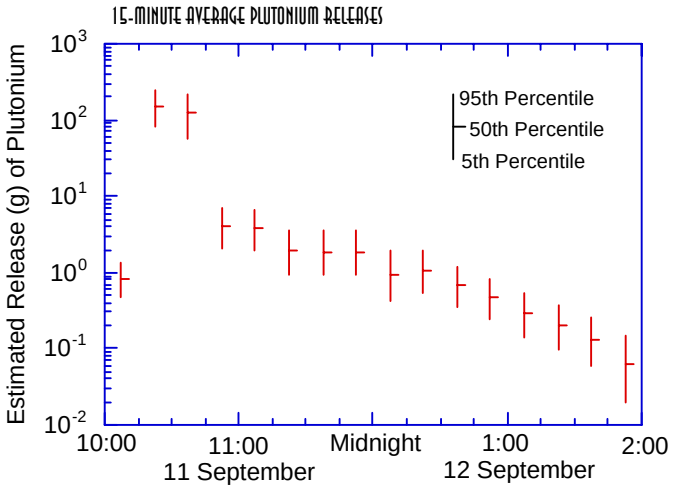

Figure S-2. Estimated releases of plutonium during consecutive 15-minute periods during the 1957 fire.

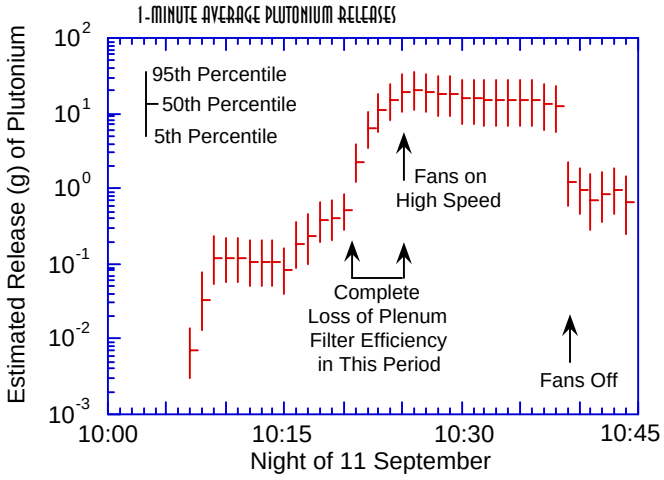

Figure S-3. Estimated releases of plutonium to the atmosphere during consecutive 1-minute periods during the first 45 minutes of the 1957 fire.

The uncertainties in the release estimates shown in Figures S-2 and S-3 reflect variations in parameters used in the calculations, including variations in release fractions for various conditions and uncertainties in the amounts of plutonium available in various forms. Also contributing are uncertainties in the amounts of plutonium previously deposited on the booster system filters, main plenum filters, glovebox exhaust filters, and room air prefilters. Those quantities involve estimates of the collection efficiencies of the various filtration systems and the historic measurements of activity in plant exhaust systems.

The release estimates discussed above reflect an assumption that the plutonium metal was evenly distributed among four categories of oxidation conditions that may have occurred during the fire. The estimated total release under those conditions was ~300 g with 5th to 95th percentile confidence bounds of 160–490 g. However, it is not certain that "vigorous" oxidation at high temperatures (>1500°C) occurred during the fire. The release estimate is most sensitive to the amount of plutonium assumed to have been in that category. Consideration of other scenarios led to the conclusion that a better estimate of the uncertainty associated with the total release is 40–500 g. The upper bound was rounded to one significant figure.

The size distributions of plutonium particles that were released at various times during the 1957 fire are not known. No measurements of the particle size of the airborne activity were performed. Experiments in which plutonium was oxidized and contaminated materials were burned have yielded a broad range of aerosol sizes. Particles with aerodynamic diameters that exceed 10 µm cannot penetrate into the pulmonary region of the lung and are not considered respirable. That diameter represents the upper bound for the sizes of particles that contribute most to dose from inhalation. For the types of plutonium oxidation and combustion of contaminated materials that occurred during the 1957 fire, consideration of a range of activity median aerodynamic diameters between 1 µm and 10 µm is recommended.

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ACRONYMS

AEC	U.S. Atomic Energy Commission
ANS	American Nuclear Society
ARF	airborne release fraction
CWS	chemical warfare system (filter)
DOE	U.S. Department of Energy
HEPA	high-efficiency particulate air (filter)
MPL	maximum permissible level
MUF	material unaccounted for
NRC	U.S. Nuclear Regulatory Commission
TBP	tributyl phosphate

ESTIMATED AIRBORNE RELEASES OF PLUTONIUM DURING THE 1957 FIRE IN BUILDING 71

1. INTRODUCTION

This investigation is a part of Phase II of the Historical Public Exposures Studies on Rocky Flats, performed by *Radiological Assessments Corporation* for the Colorado Department of Public Health and Environment. The Health Advisory Panel for the Project recommended that an independent investigation of the plutonium[a] releases from the 1957 fire be conducted as part of Task 2, Verification and Analysis of Source Terms. In Phase I, releases from the fire were estimated on the basis of environmental contamination measurements made at the time and the predicted atmospheric dispersion during the period of release. We have taken the alternative approach of using information about the material involved in the fire and the conditions before and during the fire in Building 71 (now called Building 771) to develop estimates of the amount of plutonium released to the atmosphere.

Figure 1.1 is a simple schematic diagram of portions of Building 71. The figure is not to scale and makes no attempt to show the large number of rooms and work areas in the building. Its purpose is to illustrate the primary relationships between equipment and air handling systems. These systems are important for analysis of releases from the fire that occurred in Building 71 on 11–12 September 1957. Only an overview of events is given here. A detailed chronology is provided in Section 2.

The fire started at about 10 p.m. in Room 180. The sequence of interconnected gloveboxes that housed a lathe, press, and other equipment used to form plutonium parts in that room is shown symbolically in Figure 1.1. Portions of the gloveboxes were constructed of Plexiglas.[b] The fire was initiated by spontaneous combustion of impure plutonium in a metal casting residue (called a skull) stored in the glovebox line. Subsequently, the Plexiglas ignited and burned as did the rubber gloves attached to gloveboxes.

Before the fire broke through the glovebox containment, contaminated air was carried through the glovebox exhaust lines to the glovebox exhaust filters (the so-called booster system). This system employed eight large (24 x 24 x 11.5 in.) combustible chemical warfare system (CWS) filters in two stages to remove plutonium from the glovebox exhausts. The figure shows symbolically 4 of the 12 exhausts from gloveboxes in the fire area. Although not shown in the figure, smaller CWS filters were also used at the glovebox air outlets. The fire burned through these filters and spread to the larger glovebox exhaust filters by way of the booster system exhaust lines.

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[a] In this report, the word plutonium, or its symbol (Pu), means weapons grade plutonium, which consists primarily of ^{239}Pu (~93.8%), ^{240}Pu (~5.8%), and ^{241}Pu (~0.36%). Both ^{239}Pu and ^{240}Pu emit alpha particles with average energies of 5.15 MeV and cannot be identified separately by alpha spectrometry. Releases of these two isotopes were the most important sources of radiation exposure that resulted from the 1957 fire. The beta decay of ^{241}Pu forms ^{241}Am, also an alpha-emitter, that can (after many years) account for as much as 18% of the total alpha activity. The contribution of ^{241}Am to the doses received by persons exposed during the 1957 fire was small.

[b] Plexiglas is a trade name of Rohm and Haas for several types of clear polymethylmethacrylate.

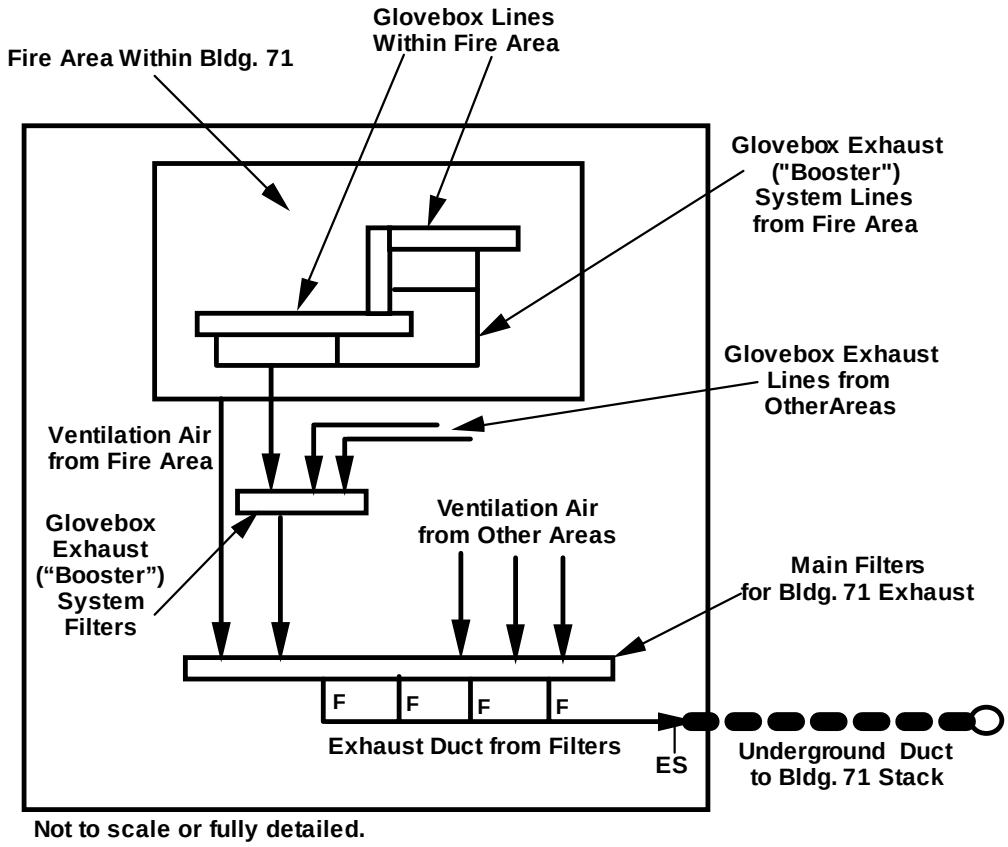

Figure 1.1. Very simplified plan view of Building 71. The glovebox exhaust system and the room air ventilation system provide air flow paths from the fire area (Room 180) to the main filter bank. The positions of exhaust fans are indicated by the letter F. The location of the effluent sampler in the exhaust duct is indicated by ES.

The glovebox exhaust filtration system discharged to the main filter plenum. The single-stage main filter bank was an array of more than 600 combustible CWS filters. These filters were the same type and size as those used in the booster system exhaust. The filter bank was also used to treat room ventilation exhaust air from many other areas in the building.

After the glovebox containment failed, contaminated air was carried to the main filter bank in the room air exhausted from the fire area. Prefilters, of the CWS type, were located at the front of the room ventilation exhaust registers. These were also burned. The fire spread to the filters in the main plenum through the glovebox and room ventilation exhausts. Exhaust fans, located on the downstream side of the main filter bank, discharged into a large metal duct that connected at the wall of the building with a concrete underground duct that carried the exhaust air to the stack.

Firefighters used water to extinguish the primary fire in Room 180 about 30 minutes after it was discovered. The fire damage on the first floor was mainly confined to that room, although connecting rooms were also heavily contaminated with plutonium. Shortly after the primary fire was extinguished, an explosion or deflagration occurred in the ventilation system. It blew contamination back through the ductwork into all parts of the building and into the atmosphere.

The exhaust fans stopped about 1 minute later because the power cable, located in the filter plenum, was burned.

Although it was subsequently necessary to attend to minor flare-ups in Room 180, the firefighters' primary focus shifted to the fire discovered in the main filter bank. That fire was declared "knocked down" at 2 a.m. The last of the smoldering filters was considered to have been extinguished by about 11:30 a.m.

The effluent sampler for Building 71 was located in the exhaust duct, near the point where it leaves the building en route to the stack. The power failure noted above precluded collecting an adequate sample of the effluent during the accident. Air sampling capability was not restored until 1 week after the fire. Failure of the effluent sampler during the accident is the reason that an alternative method is used to estimate plutonium releases from the fire.

Figure 1.2 illustrates the two pathways for plutonium release during the course of the fire. The first pathway begins at the top left of the figure. Some plutonium involved in the fire became airborne in the gloveboxes. That material was first carried to the glovebox outlet filters. When those small filters caught fire and burned through, airborne plutonium was carried to the filters of the booster system. Plutonium that had previously been deposited in the small, low-flow rate, booster system lines may have been resuspended during the fire and also carried along the same path. When the booster system filters burned and subsequently failed, plutonium was carried by the air stream directly from the gloveboxes to the main plenum filters. Part of the plutonium that had previously been deposited on the booster system filters became airborne when those filters burned. The main plenum filters had also previously been contaminated by airborne activity from various areas of Building 71.

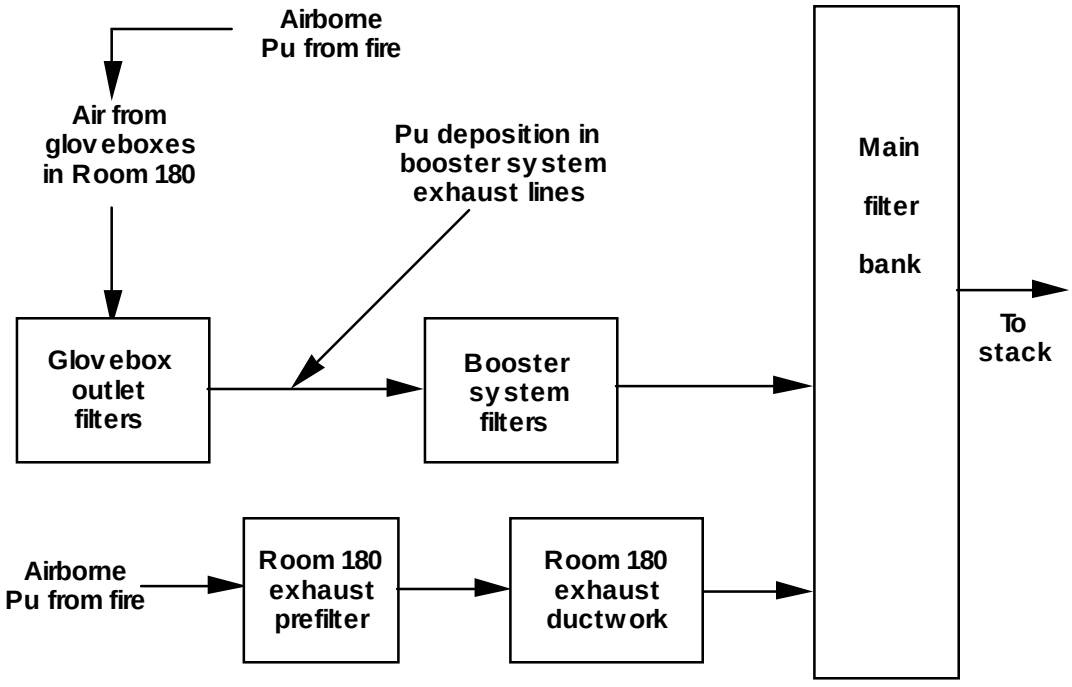

Figure 1.2. Pathways for release of plutonium during the 1957 fire.

The second pathway begins at the lower left part of Figure 1.2. When the Plexiglas gloveboxes burned through, plutonium oxidized in the fire escaped into the air in Room 180. It was carried to the inlet registers of the room ventilation exhaust and then via the exhaust ductwork to the main filter plenum. The room air exhaust registers contained prefilters that burned. Initially the filters in the plenum were intact, but they also caught fire and burned. When the plenum filters burned through, there was a direct path from the fire in Room 180 to the stack and then to the atmosphere. In addition, part of the plutonium that had been deposited on filters that burned was also released to the stack and to the environment.

Figure 1.2 indicates that information about a number of quantities, times, and parameters is needed to estimate the release of plutonium to the atmosphere during the fire. The main items are summarized below. The list also includes two items related to the explosion or deflagration that occurred in the ventilation system.

- Amount of plutonium that was involved in the fire
- Amounts of plutonium that had been collected on the plenum filters, booster system filters, other filters, and in the booster system line before the fire
- Time at which the booster system filters failed because of combustion of the filters
- Time at which the plenum filters failed because of combustion of the filters
- Number of plenum filters that were burned through
- Fraction of the amount of plutonium in the fire area that became airborne during the fire
- Fractions of the amounts of plutonium in various filters that became airborne when those filters burned through
- Cause of the explosion or deflagration in the ventilation system
- Effects of the explosion or deflagration.

The remainder of this report is devoted to collecting information about the amounts of plutonium of interest, analyzing the critical times and release fractions, and estimating the plutonium release from the 1957 fire.

Section 2 contains results of the fire investigation performed by the U.S. Atomic Energy Commission (AEC) in 1957. That report provided the basic chronology of events. Recent investigations, performed as part of the Phase II work, are also discussed in Section 2. Section 2 also presents additions to the chronology and information related to the spread of the fire. Section 3 addresses the questions dealing with sources of plutonium that was released, including prior contamination of the filter systems. Section 4 discusses release fractions that have been recommended for a variety of plutonium fires. Section 5 brings together all of the information needed to estimate releases to the atmosphere during the course of the fire. The Monte Carlo analysis of fire releases is described in Section 5. That approach reflects the uncertainties in the answers to the many questions about the fire. Section 6 presents our results and a comparison of our estimates and those made previously. Section 7 contains references. Details related to plutonium accountability and historic data on plutonium in ventilation system exhausts and in room air are provided in the appendices.

2. INVESTIGATIONS OF THE FIRE

Two sets of investigations are discussed in this section. The first investigation was conducted soon after the fire by an AEC investigation team. Their main report (Epp et al. 1957) has been declassified, with deletions of information about the masses of weapons parts. A supplementary report for that investigation (Epp 1957) was prepared later in the year. Both those reports are summarized in Section 2.1. Section 2.2 contains results of a recent investigation of the fire protection aspects. That investigation (Diliberto 1999) was undertaken to try to define important times in the sequence of events.

2.1 The 1957 Investigation Reports

Following the 1957 fire, an investigation committee was formed by the AEC. The committee completed its *Report of Investigation of Serious Incident in Building 71 on September 11, 1957* in early October (Epp et al. 1957). That report presents the basic sequence of events that occurred during the fire; factors that contributed to the severity of the fire; an estimate of the economic loss caused by the fire; and recommendations for improvements in equipment, design, and operations.

Room 180, where the fire began, was called the Development Laboratory of Building 71. It adjoined Room 179 and some small offices. The part of the room where the fire occurred was used by the Special Products Group, which produced nuclear weapons components made from alpha-phase plutonium metal. Most of the operational experience in Building 71 was related to delta-phase plutonium, a less reactive (but still pyrophoric) metal. The reactivity of alpha-phase plutonium was considered a significant factor in starting the fire. New facilities for this activity had been constructed and installed in Room 180 earlier in 1957. Some of the gloveboxes involved in the fire were constructed almost entirely of Plexiglas.

Significant events in the chronology of the fire are listed in Table 2.1. The events listed are considered most important for the purposes of estimating releases from the fire. When the fire was discovered, flames from burning rubber gloves and Plexiglas were observed. The Fire Department was called and they arrived quickly (10:12 p.m.). The 12-minute delay before active firefighting began resulted from the need to don protective clothing and respiratory protection. The fans were turned on high speed to increase ventilation of the fire area and reduce the level of airborne contaminants to which those fighting the fire would be exposed. Water was not used to fight the fire initially because of concerns about nuclear criticality, but it was employed after it became clear that using carbon dioxide had little effect. The explosion, which did not occur in Room 180, knocked down personnel in that room and in the adjacent hallway. Kennedy and Kennedy (1990) indicates that a pressure of 1 lb in.$^{-2}$ is required to produce that result. Epp et al. (1957) suggested that the explosion was probably caused by ignition of unburned gases that accumulated in the ventilation system.

The fourth item in the chronology shown in Table 2.1 indicates that the plenum filters were burning before 10:28 p.m. None of the other elements of the chronology assembled by Epp et al. (1957) provides information about the filter failure times needed for the present investigation. The absence of information about the times when filters burned through was found to be a major source of uncertainty in initial estimates of the fire releases.

Table 2.1. Elements of Chronology of Building 71 Fire (from Epp et al. 1957)

Evening of 11 September 1957	Event
10:10	Fire discovered in Room 180 by guards on a routine building tour
10:25	Building exhaust fans turned on high speed
10:24-10:27	Attempts to put fire out using carbon dioxide extinguishers failed
10:28	Smoke noticed coming from exhaust fan system
10:37–10:38	Water used to extinguish the fire in Room 180
10:39	Explosion in ventilation exhaust system
10:40	Exhaust fans went off; power cable in plenum burned through
10:47	Plenum filter bank observed to be burning
11:10	Electrical power failure in entire building
11:15	Water applied to filter fire in main plenum
Morning of 12 September 1957	
2:00	Fire in filter plenum declared “knocked down”
11:28	Filter plenum fire declared completely out

Two key factors affected the severity of the fire: (1) the use of Plexiglas in glovebox construction and (2) the use of combustible CWS filters for filtration in several locations, including the glovebox exhaust (booster) system and main filter plenum. Delays in fighting the fire were also cited. Another factor was the earlier decision to disconnect a fire detection system in the plenum area because it was the source of false alarms and unnecessary shutdowns of the building ventilation system (Epp et al. 1957).

A supplementary report (Epp 1957) prepared in December discusses factors affecting the fire and considers alternative materials for glovebox construction. Epp (1957) indicates that the flammability of Plexiglas and its heat of combustion were not fully appreciated at the time the gloveboxes in Room 180 were built. The report also addresses plant protection and fire protection improvements that were either (a) already completed or planned or (b) being contemplated. It describes reorganization of the Fire Department and discusses alternative methods to extinguish pyrophoric metal fires, which were being examined at the time. An updated cost estimate for the fire, about $430,000, was included in the report. That estimate was about 10% higher than the initial estimate that had been made.

Epp (1957) also discussed possible causes of the explosion. He concluded that it occurred in the ventilation system and was not a direct result of extinguishing the fire in Room 180 (water reacts with metallic plutonium to produce hydrogen). He noted that ventilation exhaust ductwork in rooms was not distorted and that there was no evidence of flames reaching the rooms. Distortion of ducts was found near the main plenum, and it was concluded that gases distilled from the burning CWS filters were ignited. The event was described as a “puff” or “whoosh” (Epp 1957; Epp et al. 1957) rather than a “bang.” This suggests that it was a deflagration rather than an explosion. The difference between the two types of events is primarily in the rate of burning of the flammable material and the magnitude of the pressure pulse produced (Noon 1995). A deflagration burns more slowly and produces a lower pressure pulse than an explosion,

but is not a trivial event. Persons standing in Room 180 at the time were knocked down by the resultant pressure pulse.

The fire was the subject of a serious accident bulletin (AEC 1957) in November. Building 71 was not identified and Rocky Flats was not mentioned, probably because processing of plutonium at Rocky Flats was not discussed publicly at that time. The complexities of fighting fires involving plutonium were emphasized in the bulletin, as were problems with flammable filter banks. The large economic loss sustained resulting from the spontaneous combustion of a small amount of plutonium was also noted (AEC 1957).

2.2 The Dose Reconstruction Project Fire Protection Investigation

The behavior of the CWS filters during the course of the accident is an important factor in the analysis of the plutonium releases to the atmosphere. The time required for the filters to burn through, affects the amount of time that a direct path was available for airborne plutonium to travel from Room 180 to the atmosphere. Combustion of the filters was also implicated as a source of flammable gases involved in the deflagration. How those gases could accumulate with the ventilation system operating at high speed was another puzzling question.

At the request of the Health Advisory Panel and the public, a fire protection review of information related to the 1957 fire was initiated to attempt to resolve some of these questions and to provide information needed for estimating plutonium releases to the atmosphere during the fire. The review, conducted by Diliberto + Associates, examined the original fire reports, standards and guides that were current at the time of construction and early operation of Building 71. They also reviewed publications that documented the behavior of CWS filters and other materials in fire tests.

The remainder of this section summarizes the findings of that investigation and other relevant information. Section 2.2.1 contains information about burning of CWS filters from experimental programs at Rocky Flats and elsewhere. Section 2.2.2 describes the results of testing of the flammability of other materials that were used at Rocky Flats. Section 2.2.3 provides a supplemental chronology of the fire that focuses on times that were important to estimating the plutonium release to the atmosphere. Section 2.2.4 discusses the explosion or deflagration.

2.2.1 Burning of Chemical Warfare System Filters

The combustible CWS filters were known to be a fire hazard before the fire at Rocky Flats in September 1957. The filter medium was 86% cellulose and 14% asbestos fibers, added to strengthen the paper and improve its filtration properties. The filter media and asbestos paper separators were enclosed in square plywood frames and attached to those frames using an adhesive.

In 1955, the AEC issued a serious accident bulletin (AEC 1955) that described a fire in a large bank of CWS filters in wooden frames in an Oak Ridge facility. Because water could react with the material (uranium) collected by the filters, carbon dioxide had been used to attempt to extinguish a fire in the filters. After it was believed to be out, the fire recurred on 2 successive days, again requiring large volumes of carbon dioxide for control. Subsequently, the combustible filter assemblies were replaced by a newer design that employed fiberglass filter media, metal

frames, and metal separators. The bulletin encouraged reappraisal of fire hazards at such facilities (AEC 1955).

Personnel at Rocky Flats were aware of the fire risks associated with use of the CWS filters. However, they were quite satisfied with the performance and economics of those filters. Little maintenance had been required; for example, most of the plenum filters in Building 71 had been used for 4 years. They were concerned about the possible shorter filter life and associated costs of noncombustible filters that were being designed and tested (Walker 1957). In fact, when new noncombustible filters were installed following the 1957 fire, there were a number of operational difficulties caused by poor quality assurance in filter production and short operating lifetimes (Walker 1959). An earlier presentation that described the ventilation and filtration systems in Rocky Flats facilities also suggests that cost was an important concern (Walker 1954).

Following the 1957 fire, tests of burning of the CWS filters were conducted at Rocky Flats (Erickson and Linck 1958). Earlier tests that employed the same filters had been conducted at Hanford (Keigher 1956), at the Naval War School in Virginia, and at Oak Ridge (Gilbert 1998). Diliberto (1999) reviewed the relevant tests and summarized the results. In the 1957 tests at Rocky Flats, the time required for filters to burn through was about 2.5 minutes for a single-filter flow rate of 480 ft^3 min^{-1}. That flow rate was comparable to the average flow rate through each plenum filter during the fire. The Hanford tests focused more on the temperature increase during burning. In tests with single filter flow rates of about 350 ft^3 min^{-1}, temperatures of 1100–1500°F were recorded (Keigher 1956).

A second set of tests was conducted at Rocky Flats using both CWS filters and the newer noncombustible filters. For the combustible CWS filters, single filter flow rates ranged from 224 to 525 ft^3 min^{-1}. Temperatures of about 1000°F were reached and the times required for the filters to burn through ranged from ~1 to 2 minutes (Erickson et al. 1961). The effect of flow rate was not clearly shown in the seven tests with CWS filters. Filter ignition temperatures for both the Hanford and Rocky Flats tests were in the range of 420–490°F. The adhesive used in filter construction appeared to be the first material to begin burning at a temperature of approximately 300°F.

Gilbert (1998) verbally reported his observations during CWS filter burning tests conducted in the 1950s at the Naval War School and at Oak Ridge. The following is a summary. When the filters are ignited, unburned carbon monoxide is drawn toward the back of the filter where it accumulates. When flames reach the pocket of combustible carbon monoxide, a minor explosion occurs. Gilbert observed tests that produced flame flashbacks up to 4 ft in length. Minor explosions and flashbacks were also observed in the Rocky Flats and Hanford filter burning tests (Diliberto 1999). The flashback phenomenon is important to the understanding of the postulated mechanism for the explosion, discussed later. Plugging of the filters by smoke was studied later by Gaskill et al. (1977). The filter plugging phenomenon that they observed could facilitate formation of “pockets” of carbon monoxide that are the source of flashbacks.

2.2.2 Combustibility of Other Materials

Following the 1969 fire in Buildings 776-777, Beltz et al. (1970) conducted additional tests of combustible materials used in gloveboxes and elsewhere at Rocky Flats. The tests of glovebox window material included tests of two types of Plexiglas. Gasket materials and various types of gloves were also tested. The study included combustion tests of various materials used for shielding against neutrons, a variety of paints, and components of the high-efficiency particulate air (HEPA) filters in use at the time.

Tests showed that Plexiglas began to decompose at about 390°F. The vapor that was produced was monomeric methylmethacrylate. It was quite flammable at about 540°F. Leaded gloves, composed of three layers (neoprene, lead, and neoprene), were found to be self-extinguishing at ambient temperature, but highly combustible above 390°F.

2.2.3 Supplemented Chronology for the 1957 Fire

Diliberto (1999) estimated the times of occurrence of various events important for the plutonium release calculations using information that was obtained in the burning tests described above. These times, shown in *italics*, have been incorporated into a supplemented chronology of the fire, which is provided in Table 2.2. Table 2.2 includes the elements of the chronology developed by Epp et al. (1957) that were presented in Table 2.1. A key feature of this chronology is the rapid spread of the fire from the gloveboxes in Room 180 to the booster filters and to the main filter plenum. Once ignited, both the Plexiglas glovebox material and the CWS filters burned rapidly.

The exhaust airflow in the booster system was about 4800 ft^3 min^{-1}. The flow was distributed over a square array of four filters, meaning that the flow through each filter was about 1200 ft^3 min^{-1}. Thus, the linear velocity of air through those filters was about 2.5 times higher that the air velocities employed in the filter burning tests described above. That higher air velocity is expected to have shortened the time required for the fire to burn through the two layers of booster system filters. It is estimated that at 10:12 p.m., soon after the fire was discovered, the booster system exhaust filters were compromised and provided little filtration of the air that was being carried through the booster system exhausts from Room 180.

The general arrangement of the main exhaust plenum on the second floor of Building 71 is shown schematically in Figure 2.1. This plan view is incomplete and not to scale. Its purpose is to illustrate the entry of air from various parts of the building through many (17) ducts that penetrate the concrete walls of the plenum room. The main filter bank is located in the middle of the space; air is drawn through the filters by the fans (indicated by F) and exhausted to the stack. The plenum area is very large, with an east-west dimension greater than 200 ft (north is toward the top of the figure). Ducts carrying air from the booster system exhaust and from room 180 are located on the western side of the plenum. The fire in the main plenum filters most likely started in the area where the booster system exhaust duct enters the plenum.

Table 2.2. Supplemented Chronology[a] of Building 71 Fire (Diliberto 1999)

Evening of 11 September 1957	Event
10:06	*Open flame in glovebox*
10:07–10:09	*Glovebox outlet filter burned; booster system filters catch fire*
10:10	Fire discovered in Room 180 by guards on a routine building tour
10:12	*Hole burned in booster exhaust system filters*
10:17–10:19	*Local ventilation system exhaust prefilters burning*
10:18–10:24	*Heat and smoke buildup in exhaust plenum*
10:20	*Main plenum filters begin to ignite (450°F)*
10:25	Building exhaust fans turned on high speed
10:24-10:27	Attempts to put fire out using carbon dioxide extinguishers failed
10:28	Smoke noticed coming from exhaust fan system
10:29	*Booster system filters consumed by the fire*
10:30	*Plenum filters burning*
10:37–10:38	Water used to extinguish the fire in Room 180
10:39	Explosion in ventilation exhaust system
10:40	Exhaust fans went off; power cable in plenum burned through
10:47	Plenum filter bank observed to be burning
11:10	Electrical power failure in entire building
11:15	Water applied to filter fire in main plenum
Morning of 12 September 1957	
2:00	Fire in filter plenum declared “knocked down”
11:28	Filter plenum fire declared completely out

[a] Additions to the original chronology are shown in italics.

Figure 2.2 shows the estimated spread of the fire in the main exhaust plenum. Each of the squares is symbolic of a filter in the plenum. Six filters across from the booster system exhaust duct are believed to have been first to catch fire at 10:20 p.m. The entire filter bank, which contained more than 600 filters, cannot be shown legibly in the figure. Experimental results from large-scale filter burning experiments conducted at Rocky Flats indicate that the upward spread of the fire on the filter surface would be more rapid that either the downward or lateral spread (Erickson et al. 1961). Results from those tests underlie these estimates.

An average filter dust loading of 6–10 lb per filter in plants at Rocky Flats was reported by Walker (1957) less than 3 months before the Building 71 fire occurred. He also indicated that the dust loading both raised the amount of combustible material available and contributed to the potential for spread of a fire from filter to filter.

The filters adjacent to the six that are highlighted are also estimated to have caught fire within 5 minutes after the filter fire began. The lateral spread of the fire is indicated by the times in Figure 2.2 and is considered to be symmetric; spread to the right (east) is only partly shown. The growth of the fire shown in Figure 2.2 is consistent with observations of flames on the downstream side of the filters (the side nearest the fans in Figure 2.1) between 11:13 and 11:15 p.m. (Epp et al. 1957; Diliberto 1999). Expansion of the fire area is shown for a period of

about 2 hours, from the start of the main plenum filter fire until 1 hour after the application of water to the fire began.

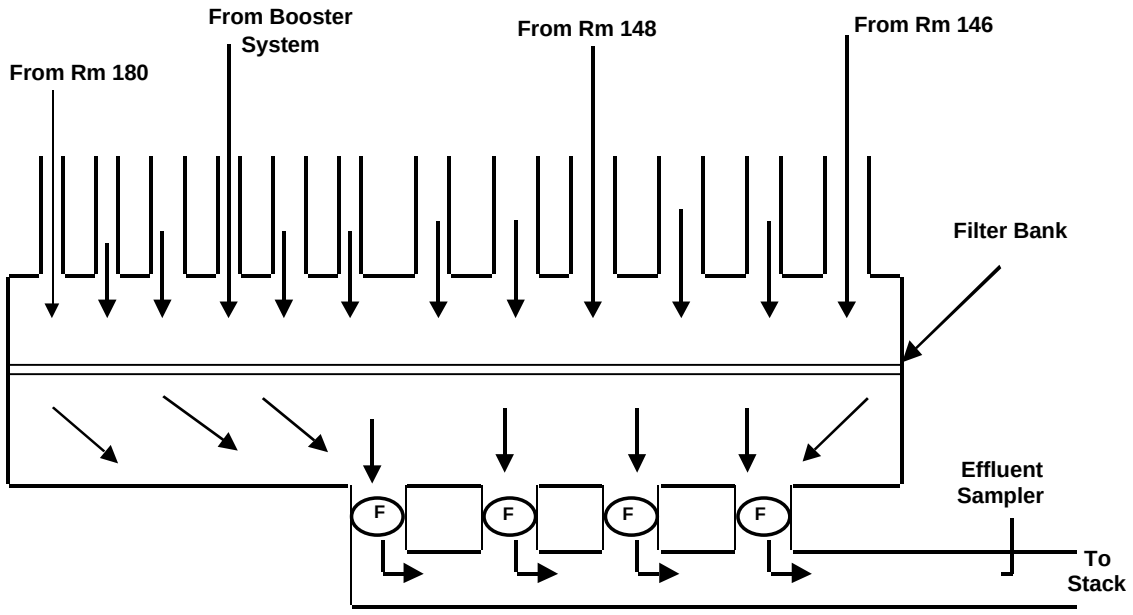

Figure 2.1. Schematic diagram of the main exhaust plenum in Building 71. Air from all parts of the building entered the plenum through ducts at the top of the drawing. The air was pulled through the filter bank and discharged to the stack by the exhaust fans.

Figure 2.3 shows the number of filters estimated to have burned through as a function of time, in 5-minute intervals, after 10 p.m. The plot reflects the fire spread shown in Figure 2.2 and the fire propagation rates from experimental tests. The first filters to catch fire are estimated to have burned through in about 4 minutes because the exhaust airflow was lower before 10:25 p.m. The rapid increase in filter failure during the 15-minute period when the fans were on high speed, before power was lost, is shown in the figure. It is estimated that filters catching fire during this period would be penetrated by fire in about 2 minutes. Most of the plutonium release (discussed in Section 5) occurred during this part of the fire.

After electrical power to the exhaust fans was lost (10:40 p.m.), it is estimated that, initially, fire burned through filters in about 5 minutes. After water was applied to the fire at 11:15 p.m., the time required to burn through the filters was estimated to be about 10 minutes. That change causes the small jog in plot in Figure 2.3; there is a 5-minute period during which no filters are estimated to have burned through. The estimated lateral spread of the fire in the filter bank, illustrated in Figure 2.2, does not change greatly during this time. The rates of fire spreading and filter burning at times after power to the exhaust fans was lost are not significant factors in estimating the overall release of plutonium.

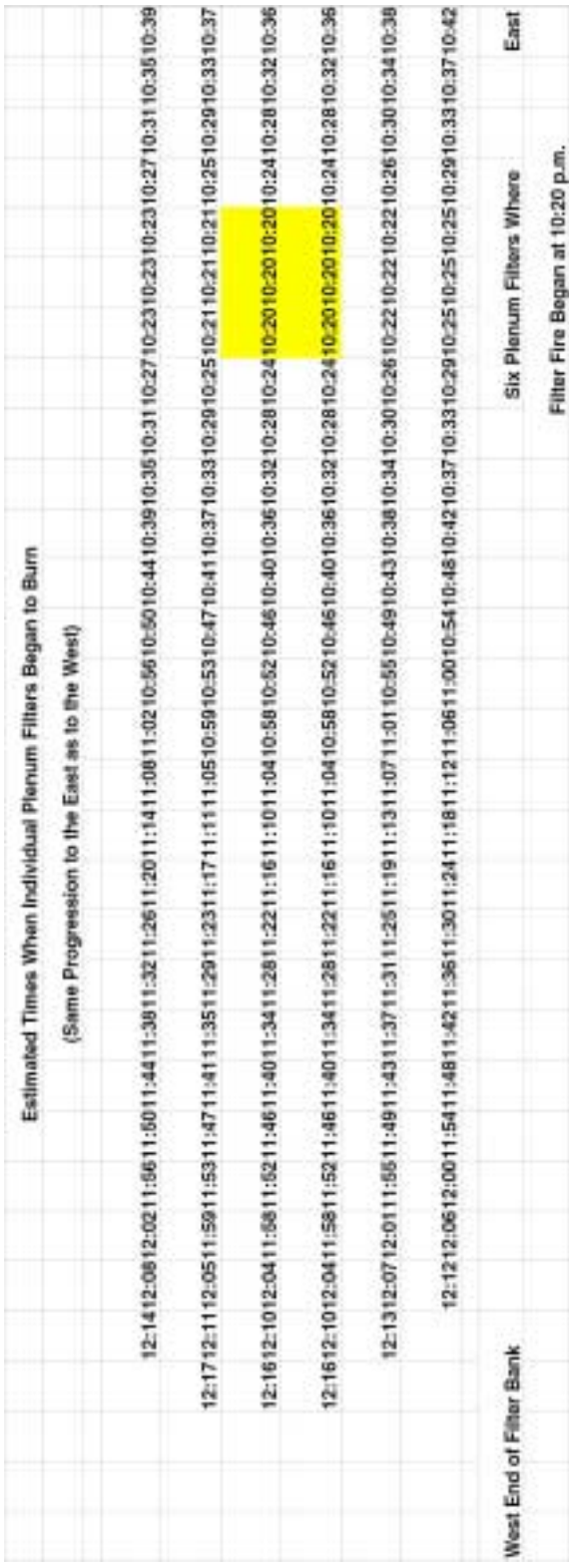

Figure 2.2. Estimated spread of the fire in the main filter plenum, 11–12 September 1957. Estimated times when filters began to burn are shown. The eastward progression is the same as the westward spread, but can't be shown. View is from inlet side of plenum.

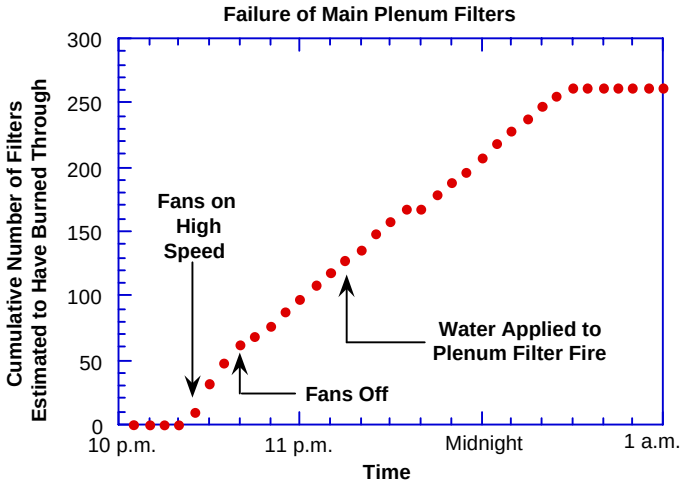

Figure 2.3. Estimated rate at which the fire burned through filters in the main plenum of Building 71 the night of 11–12 September 1957. Before 10:25 p.m., the exhaust fans operated at low speed.

2.2.4 The Explosion or Deflagration that Occurred during the 1957 Fire

Explosions (detonations) are distinguished from deflagrations by differences in rate of combustion and size of the pressure pulses produced. In a deflagration, the fuel burns more slowly and produces a lower overpressure. Most accidental explosions that occur within buildings are characterized as deflagrations. These include ignitions of flammable gases and airborne dust particles (Noon 1995). Detonations are explosions in which the rate of combustion is very fast and the overpressure produced is large. Examples of fuels for such events are dynamite, nitroglycerine, and trinitrotoluene.

The initial investigations of the fire in 1957 concluded that the fuel for the explosion was most probably unburned combustible gases that had collected in the ventilation system (Epp et al. 1957; Epp 1957). Deformations of ductwork were found near the exhaust plenum but not in the rooms, and this led to the conclusion that the explosion occurred in the plenum area (Epp 1957). There was also no mention in the 1957 reports (Epp et al. 1957; Epp 1957) of either (a) observations of a flash or (b) scorching of clothing worn by personnel who were in Room 180 when the event occurred.

Kennedy and Kennedy (1985, 1990), and Noon (1995) provide tables of the concentrations of various fuels in air that can produce deflagrations. The ranges of mixtures of fuels in air that can produce deflagrations are called the “explosive limits” for the fuels. The highest overpressure and highest flame temperature are usually achieved when the mixture is approximately chemically balanced (stoichiometric). An excess either of fuel or of oxygen can significantly reduce the overpressure and flame temperature.

The Building 71 ventilation system was reported by Epp et al. (1957) to have been operating at high speed before the event (see Table 2.1). It is, therefore, difficult to visualize a mechanism that would produce adequate concentrations of unburned gases for a deflagration to occur in the filter plenum. Although some filters may have been partially plugged by the smoke (shown in studies by Gaskill et al. [1977]), there were more than 600 filters in the plenum. Each was rated for an airflow of 1000 ft^3 min^{-1}, well above the average flow rate per filter that would occur when the fans were operated on high speed. With the fans set on high speed, the exhaust airflow was about 300,000 ft^3 min^{-1} (Walker 1954). This corresponds to the individual filter flow rate test condition (480 ft^3 min^{-1}) used by Erickson and Linck (1958). Methane and carbon monoxide are combustible gases that were likely present in the air. They have lower and upper explosive limits (volume of fuel per volume of air, expressed as a percentage) of 5% and 15% and 12.5% and 74%, respectively (Noon 1995). The lower and upper explosive limits for hydrogen, which could have been produced by the reaction of water with plutonium metal, are 4% and 75% (Kennedy and Kennedy 1985). Continuous production of large amounts of flammable gases is required to achieve volumetric concentrations of 4% or more in an airstream flowing at 300,000 ft^3 min^{-1}.

Diliberto (1999) concluded that the fuel for the explosion or deflagration was provided by the combustible paper filter media and the combustible dust and lint reported by Walker (1957) to have accumulated on plenum filters at Rocky Flats. Smoldering of this material would produce carbon monoxide. Minor explosions of carbon monoxide of the type observed by Gilbert (1998) during CWS filter burn tests, would have caused vibrations of the plenum filters. The vibrations would have suspended lint and dust and made rapid burning of it possible. The burning plenum filters provided a ready ignition source. The mechanism of flame flashbacks identified by Diliberto (1999) is more convincing than the earlier conclusion (Epp et al. 1957; Epp 1957) that invoked a simple build-up of unidentified combustible gases in the plenum.

Kennedy and Kennedy (1985, 1990) give a minimum explosive concentration for cotton lint of 0.5 ounce per cubic foot (0.0005 g cm^{-3}). Maximum explosive limits for dusts are difficult to define (Kennedy and Kennedy 1990; Noon 1995). Cotton lint is considered a “weak” explosion hazard, according to criteria developed by the Bureau of Mines, and produces a maximum overpressure of 48 psi. The maximum rate of pressure rise is 150 psi s^{-1}. (For comparison, the maximum overpressures produced by wheat and corn cob dust explosions are 103 and 110 psi, respectively. The maximum rates of pressure rise for those fuels are 3600 and 5000 psi s^{-1}, respectively, and they are rated as “strong” and “severe” explosion hazards, respectively.) All the parameter values listed and the hazard rankings are from Kennedy and Kennedy (1985, 1990).

The mass of combustible lint and dust on the plenum filters is estimated using the filter dust loading information presented for Rocky Flats. Walker (1957) reported a typical range of 6–10 lb of combustible material per filter and a maximum of 13 lb collected on some filters. Using a central value of 8 lb per filter, the total mass of combustible lint and dust is estimated to be about 5000 lb (about 2.2×10^6 g). If this mass of lint and dust were uniformly distributed within the volume (about 9.0×10^9 cm^3) on the upstream side of the filters (see Figure 2.1), the average concentration would be about 0.002 g cm^{-3}, which is about 5 times the minimum explosive concentration cited above. However, it is more likely that dust suspended from filter surfaces as the result of fire flashback explosions (Gilbert 1998) entered a smaller volume close to the face of the filter bank and was ignited.

It is known that persons in Room 180 were knocked down by the pressure pulse that resulted from the explosion. An overpressure of 1 lb $in.^{-2}$ is required to achieve this effect (Noon 1995).

Diliberto (1999) has estimated the amount of fuel that would be needed to produce such a pressure in Room 180. The calculation is not exact, but the estimated amounts range, in round numbers, from 50–70 lb of cellulosic material. It is clear, from the calculation above, that there was an abundant fuel supply available in and on the filters. Suspension of 25% of the dust collected on 30 filters would provide 60 lb of fuel.

When it occurred, the explosion would have also consumed any combustible gases present in the filter plenum. Hydrogen gas, produced by the reaction of water with plutonium metal, may have contributed to the fuel supply. The maximum production of hydrogen gas is limited by the amount of plutonium metal available for oxidation in this manner near the end of the fire. The maximum amount of plutonium metal involved is estimated to be 18 kg (see Section 3). It is highly unlikely, considering the known course of events, that it was all oxidized by interaction with the water applied to the fire. The maximum amount of hydrogen that could have been produced is estimated to be about 142 moles, which corresponds to about 3200 L or about 112 ft^3 at standard temperature and pressure. If it were produced in 3 minutes, the ratio of the volume of hydrogen to the volume of air exhausted from Room 180 (>3500 ft^3 min^{-1}) would be less than 0.01. At the higher temperatures that occurred during the fire, the volumes of hydrogen and air would both increase, but the ratio, which is well below the lower explosive limit of 4%, would not be greatly different. The upper bound concentration of hydrogen in the plenum exhaust air would be substantially smaller, but some hydrogen may have been part of the fuel supply for the explosion.

Gilbert (1998) identified carbon monoxide produced by smoldering filters as the fuel for the filter flashback explosions. Diliberto (1999) has estimated that incomplete combustion of 1 lb of cellulose fuel would produce slightly more than 1 lb of carbon monoxide. The total cellulose content of a filter is about 25 lb. This includes 8 lb of lint and dust and 17 lb (86% of 20 lb) of filter material. If partial combustion of 80% of the cellulose occurred in 5 minutes, the average volumetric concentration in air drawn through the filter at the rate of 480 ft^3 min^{-1} would be about 11%, just below the lower explosive limit for carbon monoxide. Buildup of the gas concentration because of localized plugging of the filter by smoke from the fire could produce concentration levels consistent with the flame flashbacks that were observed during filter testing. If the carbon monoxide production rate were lower, more efficient localized plugging of the filter must have occurred because flame flashbacks did occur in burning filters and were observed (Gilbert 1998).

At times after filters in the main plenum had burned through, more of the exhaust air would flow through the burned areas because that was the path of least resistance. During the period from 10:25 to 10:39 p.m., there would have been more smoldering of filters that had caught fire and, because the oxygen supply would be reduced, a greater production of carbon monoxide. It is estimated that about 60 filters had caught fire before the explosion at 10:39 p.m. It is not simple to estimate the effects of diverted airflow resulting from the burn through of some filters. The estimates of numbers of filters that had burned through, shown in Figure 2.3, assume that the rate of filter consumption by fire did not change appreciably even though the air flow rate through filters that were burning, but not burned through, had no doubt diminished. In any case, it is estimated that at least 10 filters were not burned through and were smoldering at 10:39 p.m. Thus, there were multiple opportunities for the minor explosions of the type observed by Gilbert to suspend lint and dust from the filters to fuel the deflagration that occurred. Carbon monoxide produced by smoldering filters could also have been a supplementary fuel source.

It has been suggested that a nuclear criticality may have occurred in the filter plenum and caused the pressure pulse that was experienced by those persons in Room 180. A criticality occurs when an adequate mass of fissionable material (plutonium in this case) is brought together so that a chain of nuclear fissions can occur. Each nuclear fission releases neutrons, some of which cause further fissions. The reaction continues until the energy released separates the mass of plutonium into a configuration that does not allow a chain reaction to occur. The energy produced by the fission reactions would also cause a pressure pulse. The amount of plutonium required to produce such a reaction is known as the critical mass. It is highly dependent upon the geometric array of the plutonium components. The lowest critical mass of metal, a few kilograms, is usually found for a spherical geometry. However, the critical mass depends on the form of the material and whether neutron reflectors are present.

In addition to producing a pressure pulse, a criticality would also have produced neutron and gamma ray fluxes and fission products that would have been blown back into the building and out into the environment. However, there are no reports of elevated external exposure rates that would have been produced by the fission products or of the presence of fission products on contamination smear samples or air samples collected within the building. There are also no reports of alarms from the criticality monitors located in the processing areas on the first floor of Building 71.

Nearly all of the plutonium (mean estimate of ~300 g) that was collected by the plenum filters before the fire came from Room 146 as a result of the peroxide explosion in June 1957 (see Section 3). That exhaust duct enters the plenum near the west end (see Figure 2.1). If the plutonium from that explosion were concentrated on a few filters, it would be at that end of the plenum. If it were spread over many filters, the amount per filter would be small. In either case, the planar distribution of material would not be conducive to producing a criticality when more plutonium was added during the September 1957 fire. The amount of plutonium carried to the filter plenum during the September fire is estimated to be less than 600 g (see Section 5). Not all of this plutonium was present at one time, and it is estimated that much of it was released through the damaged filter plenum. Plutonium that was collected on surfaces of several filters would not be in a geometry that was conducive to creating a critical mass. The amounts of plutonium on the plenum filters during the fire are too small to produce a criticality. The original classified investigation documents have been reviewed and found to contain no evidence or suggestion that a nuclear criticality was the source of the pressure pulse.

2.3 Summary of the Fire Investigations

This section has focused primarily on the aspects of the fire that are important to estimating the releases. The summary that follows identifies a number of the key elements, including some that are related to fire protection. Further discussion of the fire protection aspects of the investigations and recommendations that were made can be found in the investigation reports (Epp et al. 1957; Epp 1957; Diliberto 1999). Some of the same fire protection issues resurfaced in 1969 when a second major plutonium facility fire occurred at Rocky Flats in Buildings 776-777 (see Voillequé 1999a).

The investigations of the 1957 fire have revealed a number of important factors related to the fire and its spread. The initial investigation led to experimental investigations that have proved quite useful in defining a more detailed chronology of events. The following aspects are particularly noteworthy:

- The potential for spontaneous combustion of alpha-phase plutonium was not fully appreciated; protection against a fire initiated by that mechanism was not adequate
- The use of Plexiglas in glovebox construction contributed significantly to the spread and severity of the fire
- Use of combustible CWS filters in exhaust cleanup systems allowed the fire to spread to the filter plenum; burning of CWS filters created a direct path from the fire area to the atmosphere
- Operation of the exhaust fans at high speed provided some protection to firefighters but also provided oxygen to the fire, increased the rates of filter combustion, and led to larger releases of plutonium to the environment
- Evaluation of filter burning tests that simulated conditions present during the fire permitted definition of a more detailed chronology of events important to estimation of releases
- The pressure pulse that knocked down persons in Room 180 was most likely caused by a deflagration fueled by combustible dust and lint that had accumulated on the plenum filters and by carbon monoxide (CO)
- Production and pocketing of CO in smoldering filters led to flashbacks that likely initiated the deflagration that occurred in the ventilation system
- Accumulations of combustible dust and lint on filters facilitated the spread of the fire in the main filter plenum and provided fuel for the deflagration that occurred
- Hydrogen gas produced by the reaction of water with plutonium metal was at most a minor contributor to the deflagration
- There is no evidence that a nuclear criticality occurred during the 1957 fire.

3. SOURCES OF RELEASED PLUTONIUM

Figure 1.2 shows that there were several sources of plutonium that could contribute to airborne releases during the course of the fire. Those sources are discussed in this section. The first, and most important, is the plutonium in the fire area in Room 180 (Section 3.1). Section 3.2 discusses estimates of the amounts of plutonium that had accumulated on the main plenum filters, booster system filters, glovebox outlet filters, and room air exhaust prefilters before the fire. Taken together, the amounts of plutonium collected by various filters are the second most important source of plutonium releases. Section 3.3 considers the amounts of plutonium deposited in ductwork that may have been suspended during the course of the fire. Section 3.4 provides a summary of the various quantities identified.

3.1 Plutonium in the Fire Area

The first and most important source of released plutonium considered is material that was present in the fire area and was involved in the fire. Epp et al. (1957) and Epp (1957) reported the damage that occurred in the fire area in Room 180. That information is summarized here. Figure 3.1 shows the general layout of the glovebox line in Room 180. The fire was initiated by spontaneous combustion of plutonium stored on a shelf in the west conveyer box. A Plexiglas partition between the conveyer box and the "work area" was ignited and that Plexiglas box was totally burned. The box that contained the lathe was also severely damaged. The inspection box was also partly constructed of Plexiglas and it was burned. Portions of the glovebox line were not greatly affected by the fire. The storage box windows were only deformed by the heat but not burned. Unburned rubber gloves were found in parts of the glovebox line that were north and east of the cold storage box.

The 1957 investigation report (Epp et al. 1957) provided an initial estimate of the amounts of plutonium in various parts of the room. This inventory was based upon the 3 September accountability report and interviews with workers. Although the accountability data were incomplete at the time, the report provides information on the forms and physical locations of much of the plutonium present at the time of the fire. Table 3.1 summarizes the information on the amounts of plutonium by location relative to the fire in Room 180. The fire damage was described above. The north-south line that terminates with airlocks (see Figure 3.1) was adjacent to the burned area. The eastern end of the conveyer line and the line between the mill and the "B" box are the unaffected area.

At the time of the investigation report (7 October 1957) the amount of plutonium in the room at the time of the accident was given as 42.3 kg. However, the report identifies some additional material that had been identified but whose mass was not yet known. Not included in the stated inventory were the amount of plutonium in a large bottle of degreasing solvent, the mass of some miscellaneous metal and sweepings, and the masses of three casting residues. The sum of the identified masses is 26.5 kg. The masses of the hemispherical shells of plutonium metal, which were included in the inventory, were unspecified because they are classified. Assuming that indicated uncertainties in the amounts of plutonium chips in cans are small, the average mass of the nine metal hemispheres can be estimated to be <1.8 kg.

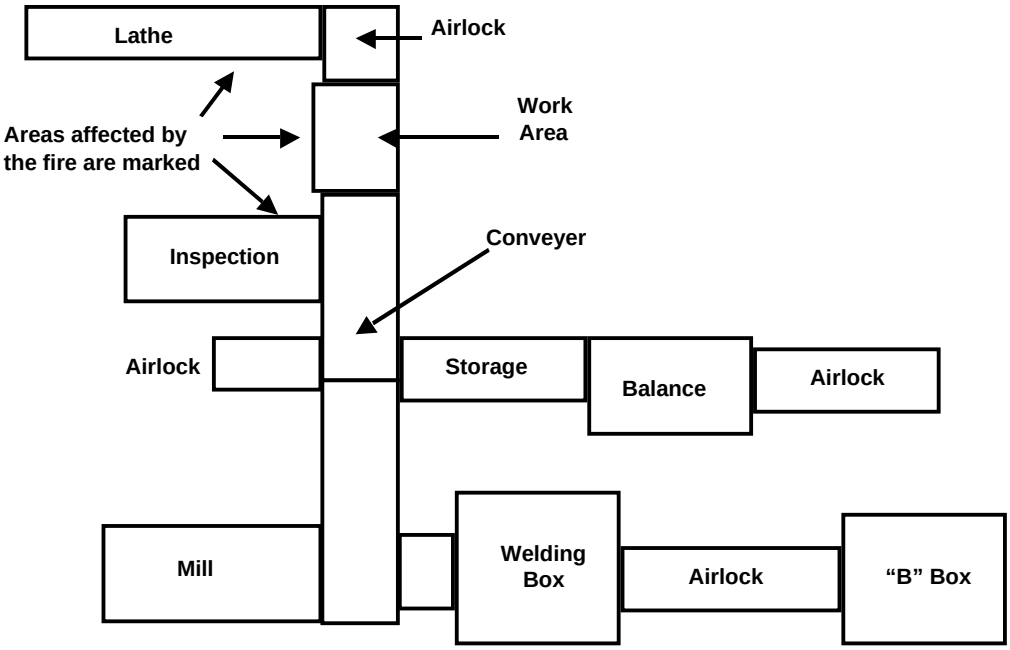

Figure 3.1. Diagram of glovebox line in Room 180 where the fire began. The shading indicates the approximate area burned by the fire.

Table 3.1. Locations of Plutonium in Glovebox Line in Room 180 Before the Fire[a]

Description	Location, by section of glovebox line		
	Area burned by fire	Adjacent to burned area	Unaffected area
	Amount of plutonium (kg)		
Plutonium in organic liquids (lathe cutting oil sludge, CCl_4 degreasing solvent)	1.2	[b]	2.1
Plutonium metal pieces of various types	0.5	1.6[c]	[d]
Plutonium metal casting residues	0.8	1.2[c]	[d]
Plutonium chips in cans	[d]	3.7[e]	8.0[e]
Plutonium dioxide (PuO_2) in cans	2.6	2.2	2.6
Totals	5.1	8.7	12.7
	Number of hemispheres[f]		
Plutonium metal hemispheres	8	[d]	1

[a] Based upon the 7 October accident investigation report (Epp et al. 1957). At that time, the known amount of plutonium in the room was estimated to be 42.3 kg.

[b] A large plastic bottle containing 8 L of carbon tetrachloride degreasing solvent with an unknown amount of plutonium was identified in this area.

[c] In this area, additional materials (consisting of miscellaneous metal, three casting residues, and sweepings) were reported, but the masses of those materials were not given.

[d] No material of this type was present.

[e] These masses were indicated to be uncertain.

[f] Masses of hemispheres were not given; average is estimated to be <1.8 kg (see text).

Nearly all the metal items that were identified as having been in Room 180 before the fire were experimental alpha-phase plutonium, as opposed to the delta-phase plutonium handled in larger quantities elsewhere in Building 71. The work in Room 180, the special products area, was related to developing new nuclear weapons. Inexperience with alpha-phase plutonium was considered a contributing factor to the occurrence of the fire (Epp et al. 1957).

The amount of plutonium in the 8-L bottle of degreasing solvent is not expected to be large. If the plutonium concentration in CCl_4 were 100 times that found in waste solvent and if the bottle were full, then only about 8 g of plutonium would have been present. The amount of plutonium reported to have been present in lathe cutting oil and sludge has been rounded from 1170 g to 1.2 kg. Use of 1.2 kg with a substantial uncertainty (see below) accounts for the amount of plutonium that was in the 8-L bottle.

The plutonium accountability procedures were applied to the fire area until cleanup of Room 180 was completed in 1961. Those accountability data, which remain classified, have been reviewed and the elements useful for this analysis have been extracted. Our notes containing that information have been declassified. Appendix A discusses the accountability data related to the 1957 fire and the estimates of the amount of material unaccounted for (MUF) following the fire. The U.S. Department of Energy estimated loss of plutonium resulting from the fire to be 6 kg (DOE 1994a) based on analysis by Zodtner and Rogers (1964). Our analysis of the accountability reports indicates that the total amount of material that was unaccounted for following the fire was probably 8.3 kg (see Appendix A).

The following discussion digresses from the topic at hand, but it is closely related to the plutonium accountability information used in this section. Initially, it was thought that the accountability data could provide a basis for estimating the amount of plutonium released during the 1957 fire. The theoretical approach is described in Appendix B. Investigation showed that the mass balance approach could not be used to make an unbiased estimate of the release because of a serious flaw in the accounting system. Before 1967, reliable measurements of the amounts of plutonium in solid waste shipments were not performed. Appendix A provides information about the quantities of solid wastes shipped and discusses the measurement difficulties. This systematic inadequacy in the plutonium accounting process prevents an estimate of the amount of MUF from being a reliable guide to the amount of plutonium released to air or water. The amount of MUF does provide a grossly overestimated upper bound on the amount of material released. Such upper bounds are useful for some types of screening calculations, but they are not credible estimates of the amounts of plutonium actually released in airborne and liquid effluents. [c]

The October and November 1957 accountability reports (Appendix A) gave differing estimates of the quantities of plutonium present before and after the fire. These changed as more information was collected. Later estimates of possible losses of plutonium were consistent with the November report, and that report is consistent with the analysis by Zodtner and Rogers (1964). The final estimate of the pre-fire inventory was 62.5 kg of plutonium. In November 1957, about 37 kg of the plutonium had been recovered from the room. An additional 16 kg had been located in the room and another 5 kg was thought to be present but had not been located at that

[c] Appendix B also includes an example of the mass balance accounting system and its relationship to estimates of routine releases to air and water from a plutonium processing facility. For a facility much like Rocky Flats, our analysis shows that the mass balance approach has no potential for determining the amounts of material released routinely. This result is consistent with the findings of a committee of the National Academy of Sciences for releases from chemical facilities under similar conditions (NAS 1990).

time. From that report, it can be estimated that about 9 kg of plutonium, including the material that had not been located, was definitely involved in the fire.

Unfortunately, there is no way to correlate the amounts of plutonium in the accountability summaries with the listing of materials in particular forms (Table 3.1) given by Epp et al. (1957). The detailed compilations of information that were used to prepare the summaries have not been retained in the plant records system.

There are two categories of material that could have been oxidized during the fire: (1) the metal turnings contained in lathe oil and sludge and (2) the various types of plutonium metal (including unidentified pieces, residues, and hemispheres). The amount of plutonium in oils and sludges is estimated to be between 1.0 and 2.4 kg. This range reflects possible measurement uncertainties and also the fact that there were uncertainties about the amounts of plutonium in sludge that may have burned (Epp et al. 1957; Epp 1957). There were eight plutonium hemispheres in the area where fire damage was greatest. An upper bound on the mass of plutonium metal in these components is 14.4 kg. Considering the other metal forms that were present leads to an upper bound metal mass of about 16 kg. A lower bound for the amount of metal oxidized is taken to be 9 kg based upon the November accountability report. In the Monte Carlo calculations in Section 5, the ranges specified here are taken to be the bounds of uniform probability distributions. The conditions of oxidation of plutonium metal are very important for estimating the appropriate release fraction (Section 4). Because these conditions are not known, a range of possibilities is considered in Section 5.

3.2 Plutonium Previously Retained by Filter Systems

This section describes the estimates of plutonium that had accumulated on filters in the ventilation system before the fire. The filters of interest include the large main plenum filter bank, the booster system filters, the glovebox outlet filters in Room 180, and the room air prefilters in Room 180. The estimates are based upon in-plant measurements of plutonium concentrations in ventilation air. Thus, knowledge of the airflow in the various parts of the ventilation system is an essential component of the procedure. Section 3.2.1 presents information on the airflow rates in the building ventilation exhaust system. Subsections that follow discuss estimates of the amounts of plutonium in the locations of interest. Section 3.2.2 deals with the main filter plenum. Section 3.2.3 discusses the amounts of plutonium collected by the glovebox exhaust system (booster system) filters. Section 3.2.4 addresses the glovebox exhaust filters and the ventilation air duct prefilters in Room 180.

3.2.1 Exhaust System Air Flow Rates

There were many ducts that carried air to the main filter bank. The general airflow in the building was from office and hallways into rooms that contained processing lines and then into the contaminated gloveboxes, which were at the lowest pressure. Air entering the gloveboxes was filtered and local exhaust filters were also provided. The collected glovebox exhausts from the analytical and development areas were carried to the booster system filters. Glovebox exhausts from the chemical, fabrication, and recovery areas were collected and filtered by the recirculating air system (so-called because it was designed to be an inert atmosphere system to which only makeup nitrogen would be added). The filtered discharges from the recirculating and booster systems were combined with room air exhausts and transported to the inlet side of the

main filter bank. After filtration, all the ventilation air was discharged via the Building 71 stack. Figure 2.1 showed the discharge of the ducts into the main filter plenum, the plenum filters, and the exhaust system.

Routine monthly reports prepared by the radiation protection staff of Building 71 contain some information about building ventilation system flow rates as well as all the data from measurements of airborne plutonium (Putzier et al. 1953–1958). Data in the January 1954 report indicate that the flow rate during the day shift was 180,000 ft^3 min^{-1}. At night and over weekends, the flow rate out of the building was lowered to 130,000 and 100,000 ft^3 min^{-1}, respectively. Air supply fan operation was adjusted accordingly. The data on fan operation were apparently kept by building operations at the time, but no detailed records of the information have been found.

The January 1954 flow rates are generally supported by information contained in an Argonne National Laboratory trip report prepared following a visit to Rocky Flats (Shuck 1954). That report states that the fans could be operated at three different speeds, normally providing 20 air changes per hour, and reduced to 10 air changes per hour in off shift hours. It was stated that, at high speed, the fans could produce 30 air changes per hour. The rates of 20 and 10 air changes per hour are roughly consistent with the daytime flow rate of 180,000 ft^3 min^{-1} and the weekend flow rate of 100,000 ft^3 min^{-1}. The higher air change rate reported by Shuck (1954) probably refers to the condition when all four fans are operated at full capacity. That interpretation is consistent with the flow rate of about 300,000 ft^3 min^{-1} estimated for fan operation at high speed during the fire (Section 2.2.4) and with the drawings of the ventilation system for Building 71 (Austin 1952).

It is known that the daily average exhaust flow rate increased as multiple shift operations became more common. There were two reasons for the increase. First, the operational flow rate was used for a greater fraction of the day. Second, the normal operational flow rate was increased to 202,000 ft^3 min^{-1}. A sheet of air duct data dated December 1955 (Anonymous 1955) shows a daytime linear velocity for the main exhaust duct of 3360 ft min^{-1}, which corresponds to that higher flow rate. The measured night time air velocity in the duct was 1700 ft min^{-1}, implying a total discharge of about 102,000 ft^3 min^{-1} under those fan operating conditions (Anonymous 1955).

Air velocity measurements were reported for four of the main exhaust ducts as well as for the main filter plenum (Anonymous 1955). Airflow rates for those ducts averaged 64% (range: 58–72%) of the flow rates shown on building drawings (Austin 1952) and are consistent with the flow rate of 202,000 ft^3 min^{-1} derived from the air velocity measurement for the main exhaust duct. The sum of the flow rates shown in the drawings for all ducts entering the filter plenum is about 312,000 ft^3 min^{-1}. This corresponds to the maximum exhaust flow rate for the building discussed above. The ratio of the measured building flow rate to the total flow rate from the drawings is 0.65.

To calculate the amount of plutonium transported to the main filter plenum by an exhaust duct, it is necessary to know the flow rate of air in the duct and the concentration of plutonium in the air stream. The measurements made in December 1955 were used as a reference point for estimating airflow rates through the ventilation exhaust ducts. The maximum flow rate specified for an exhaust duct (Austin 1952) was multiplied by 0.65 to obtain the flow rate that corresponded to operation with a total exhaust flow rate of 202,000 ft^3 min^{-1}.

Table 3.2 lists the building ventilation exhaust ducts and contains the duct flow rates that correspond to a building exhaust rate of 202,000 ft^3 min^{-1}. There has been no attempt to provide

a comprehensive description of all operations in the areas served by these ducts; however, some general information can be provided. The first five ducts listed served the major processing areas (Rooms 146, 148, 149) and glovebox exhaust systems. These ducts were sampled routinely and are discussed further in the next section. Most of the building utilities were located on the second floor as were rooms housing filtration systems. Rooms 179 and 180 were in the development area. Rooms 181–182 were used for plutonium fabrication. Plutonium components were stored in Rooms 187–188 (ERFOM 1994).

Table 3.2. Flow Rates for Ventilation Exhaust Ducts in Building 71

Duct label	Flow rate[a] (ft^3 min^{-1})	Primary rooms or areas exhausted
D–74	29,000	Room 146 and Building 74
D–77	30,000	Rooms 148, 148A, recirc. air flow
D–84	20,000	Rooms 149, 149A, and 149B
D–86 (partial)	4,800	Booster filtration system
D–88	26,000	Development and analytical areas
D–75	7,000	Second floor
D–76	8,700	Room 181 east
D–78	8,400	Room 148 (partial)
D–79	3,900	Room 171
D–80	5,800	Room 182 east
D–81	11,400	Room 182 west
D–82	2,100	Rooms 166, 174
D–83	11,200	Room 181 west
D–85	4,100	Second floor
D–86	16,800	Room 179
D–87	3,500	Room 180
D–89	10,400	Rooms 180, 185, 186
D–90	2,400	Rooms 186, 187, 188
Main exhaust	202,000	Entire building

[a] Flow rates consistent with a daytime flow rate of 202,000 ft^3 min^{-1} for the main exhaust. That average flow rate was not approached on a regular basis until mid-1956.

Original records of the exhaust flow rates during early years of operation of Building 71 are no longer available for review. However, the monthly health physics reports (Putzier et al. 1953–1958) contain discharge air volumes for the months December 1953 through April 1954. Other monthly health physics reports contained both the amount of plutonium released and the measured effluent concentration. For those months, the volume of air that was discharged from the building has been computed by taking the ratio of the amount released to the average concentration in the effluent. The results are shown in Figure 3.2. Average air discharge rates were quite variable during the first 3 years of operation and were generally below 120,000 ft^3 min^{-1}. They were consistently higher after mid-1956.

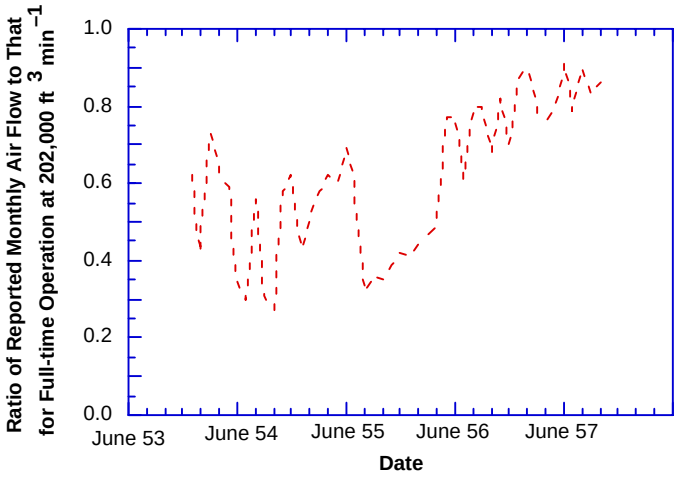

Figure 3.2. Reported or calculated monthly exhaust airflows for Building 71. Fractions of the airflow for full-time operation at 202,000 ft^3 min^{-1} are shown. That flow rate was not approached consistently until mid-1956.

Figure 3.2 shows that using the reference flow rate of 202,000 ft^3 min^{-1} to determine the flow rates for individual exhaust ducts could lead to an overestimate of the amount of plutonium carried to the main plenum filters. Before June 1956, the average ventilation flow rate was only about half of the reference value. During the latter part of 1956 and in 1957, when the most important events that caused contamination of the plenum filter bank occurred, the amount of overestimation is much smaller. For that reason, the use of reference flow rates (Table 3.2) for the individual ducts throughout the entire time period is considered to be an adequate procedure even though it will bias the results somewhat on the high side.

3.2.2 Plutonium Collected on the Main Plenum Filters

The fire in the main filter plenum burned many of the more than 600 CWS filters that were installed there. The extent of the damage to the filter bank is not quantified in historic reports. It is known that firefighters created firebreaks by using pikes to pull some of the filters out of their holders. This was done to reduce the lateral spread of the fire (Epp et al. 1957). The width of the firebreaks was not stated. At least two columns of filters must have been pulled down to make the firebreaks. That would have affected a dozen filters and is consistent with a report (Freiberg 1999) that few filters were removed. Photos of cleanup operations show that some filters remained in place, but there are no photos of the filter bank immediately after the fire.

The amount of contamination present on the filters is needed to estimate the release of plutonium from filters that burned. The plenum filter contamination from operational and accidental releases of plutonium into the ventilation systems in Building 71 before 11 September 1957 is discussed here. The contribution from the most important ventilation exhaust ducts is discussed first. These ducts carried about half of the air, primarily from processing areas, to the

main filter plenum. Then the contributions from other ducts, which were not sampled routinely, are discussed.

3.2.2.1 Filter Contamination from Primary Exhaust Feeder Ducts. Sampling of the Building 71 effluent discharge was initiated in June 1953 when operations began. At the same time, sampling of the duct that carried the exhaust from Rooms 148 and 148A and the bleed from the recirculating air system was also initiated. This was one of the most important ducts carrying air to the main filter bank. By early 1954, the sampling program had been expanded to include the five feeder ducts, which handled the ventilation exhaust air expected to contain the highest levels of contamination. Table 3.3 shows the ducts that were sampled and the functions performed in the areas that they served. Flow rates for these ducts are given in Table 3.2.

Table 3.3. Ducts in Building 71 That Were Sampled Routinely

Duct label	Areas from which air was taken	Description
D–74	Room 146 and Building 74	Plutonium and waste processing operations
D–77	Rooms 148, 148A, and bleed from recirculating air system	Plutonium processing operations, filtered exhaust air from processing, recovery, and fabrication gloveboxes
D–84	Rooms 149, 149A, and 149B	Plutonium recovery operations, incinerator
D–86	Booster exhaust filtration system	Filtered exhaust air from gloveboxes in the development and analytical areas
D–88	Development area (Rooms 180, 178–179, 174–176, and 172) and analytical area (Rooms 151–154 and 156–169)	Special products processing and fabrication operations, sample analysis support efforts

Table 3.3 shows that the duct sampling program was focused on those areas where the highest levels of airborne plutonium were expected. The gas that was filtered and bled out of the recirculating system for the gloveboxes in the main processing areas was part of the D–77 exhaust. Measurements of that exhaust alone were performed during part of the time before September 1957. The air exhausted from the main plutonium processing and recovery operations was carried through ducts D–74, D–77, and D–84. Initially, duct D–74 included the exhaust from Building 74, the waste processing facility. That exhaust was later separated, filtered, and sampled separately. The exhaust air from the development area, including Room 180, and the analytical areas was carried in duct D–88. The booster system filters treated the air exhausted from gloveboxes in these areas and it was then discharged into duct D–86. That exhaust contributed most of the plutonium activity in duct D–86.

Initially, sampling was performed only during the day shift hours, when operations were conducted. During off shifts, the exhaust flow rate was reduced because of limited activity in the building, but it was not sampled. Continuous air sampling was performed after the first year of operation. The program initially changed to collection of separate samples for the day shift and overnight period. Later, daily 24-hour samples were collected during the week and longer continuous samples were collected over weekends and holiday periods.

The duct sampling systems were designed to extract samples isokinetically. The health physics vacuum system was capable of maintaining the steady flow rates needed to achieve this goal. During the period of interest for this report, the samples were analyzed for total long lived alpha activity. The samples were counted twice and the method of Koval (1945) was used to account for the contribution of naturally occurring alpha-emitting radon progeny also collected on the sampling filter.

The routine monthly reports of the radiation protection staff in Building 71 (Putzier et al. 1953–1958) contain the results of the air sampling program for the building stack discharge to the atmosphere and for the exhaust ducts described in Table 3.3. The reported effluent and duct air sampling results are tabulated in Appendix C. Initial estimates of the amounts of plutonium carried to the plenum filters were calculated using the plutonium air concentration data listed in Appendix C and the reference flow rates for the sampled ducts (Table 3.2). Corrections of the initial estimates are discussed later in this section.

Table 3.4 shows the contributions of each flow path. The estimated amounts are presented in both activity and mass units. The initial estimate for Building 74 covers the period beginning in March 1956 when that exhaust was isolated and filtered. Separate sampling of that exhaust, whose reference flow rate was 13,000 ft^3 min^{-1}, began at that time.

Table 3.4. Initial Estimates of Inputs to the Main Filter Bank Before the Fire in Building 71 on 11 September 1957

Duct number	Initial estimates of input to main filter bank[a]	
	(µCi $^{239/240}$Pu)	(µg $^{239/240}$Pu)
D–77	5.4×10^4	7.5×10^5
D–74	1.2×10^7	1.7×10^8
Bldg. 74[b]	7.7×10^2	1.1×10^4
D–84	7.2×10^3	1.0×10^5
D–86	5.3×10^2	7.4×10^3
D–88	9.4×10^2	1.3×10^4

[a] Correction of these estimates are discussed later (see text).

[b] In March 1956, a separate filtered duct and a separate sampling point were installed for Building 74. Before that time, that effluent was carried in D–74.

The principal contributor to the filter bank activity was duct D–74, which served Room 146. An explosion occurred in that room in June 1957 (Shepherd et al. 1957) and was responsible for the vast majority of plutonium transported to the filter system. The estimated total for that exhaust, and that event, is 12 µCi (168 g) of $^{239/240}$Pu. There were a few other events whose contributions to the total could be identified. Those were (1) maintenance activities in Room 146 in early 1956, (2) bag-out operations in Room 148 in March 1956, and (3) pump repair in Room 146 in January 1957 (Putzier et al. 1953–1958). Although the results of these activities can be detected, the resulting amount of plutonium carried to the main filter plenum is estimated to be less than 1 g.

Figure 3.3 is a plot of the initial estimates of buildup of $^{239/240}$Pu on the main filter bank from startup of Building 71 operations until 11 September 1957. The dominant feature is clearly the increase in total activity resulting from the peroxide explosion in June 1957. The increases from the problems with bag-out operation in March 1956 and to the pump repair work can be discerned. Although the quantities involved in those two cases were approximately equal, they appear to be different because of the logarithmic scale of the figure. It is possible that some early contributions may have been underestimated because the sampling program was not fully implemented until early 1954. Use of the reference flow rates in the calculation generally overestimates the amounts for measured concentrations. As a result, it is expected that the effect on the total of any underestimation during the first 6 months would be quite small.

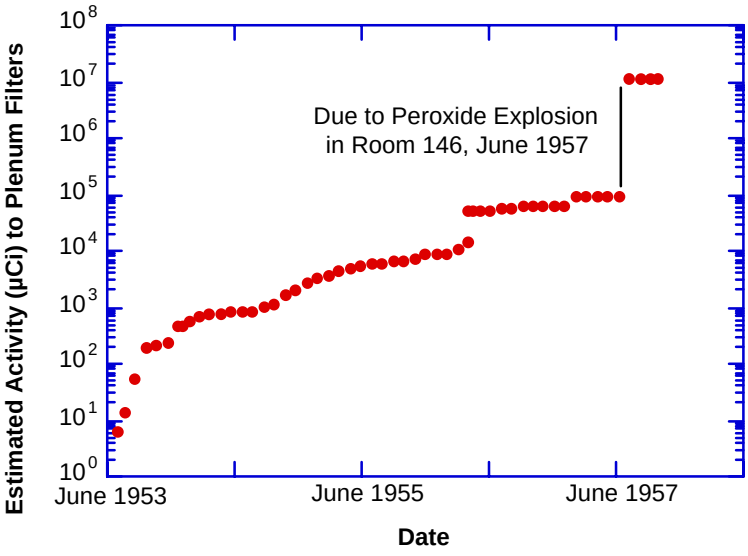

Figure 3.3. Estimated buildup of $^{239/240}$Pu activity on Building 71 main filter bank from startup until 11 September 1957. The final value of 1.2 x 10^7 µCi corresponds to a $^{239/240}$Pu mass of 1.7 x 10^8 µg.

The Building 71 incinerator, which later became a significant source of plutonium contamination of the exhaust system, was installed in Room 149 in 1957. However, it does not appear to have been operated frequently during that year. In his review of the history of Building 71, Owen (1963) does not identify incinerator problems until 1958. In March of that year, the building exhaust concentrations were about 4 times higher than in February and contamination of the duct was measured. Contamination problems caused by the incinerator in 1960–1963 were even more severe (Owen 1963).

A review of the log of special air sampling efforts (Anonymous 1957) revealed several special samples for incinerator maintenance (bag changes, filter changes) in November and December 1957. There was also an incinerator fire reported on 18 December. However, all of

these problems associated with incinerator operations in 1957 occurred after the fire in Room 180.

A deficiency of the Building 71 effluent and duct sampling program was that there was only one sampling point in each of the ducts. The ducts that were sampled were large, with cross-sectional areas of 16–21 ft^2 for the main feeder ducts and 60 ft^2 for the main exhaust duct. Sampling guidance (ANSI 1961) recommends multiple probes for such large ducts. The use of single sampling probes raised a serious question about whether the samples collected were representative of the plutonium concentrations in the various ducts.

Voillequé (1999b) reviewed sampling results for the largest duct, the main building exhaust. Single-point sampling of the main exhaust was replaced by 3-point sampling in 1963. Data collected after that time provided a basis for evaluating whether the samples from the single centerline point were representative. Analysis of the data showed that the single probe did not provide representative sampling of the building effluent, and that the likelihood that the discharge would be underestimated was greatest when effluent concentrations were low. Under all conditions there was considerable uncertainty associated with corrections for samples that were not representative.

There are no comparable multi-point sampling results for the feeder ducts described in Table 3.3. The fact that the cross-section of the main building exhaust duct is about 3 times larger than those of the feeder ducts does not necessarily imply that single-point sampling of those ducts will be more reliable. The methods developed by Voillequé (1999b) were used to estimate the sampling bias associated with the reported concentration in the Room 146 duct as a result of the peroxide explosion. The median estimate for those conditions was 1, with a geometric standard deviation of 1.5.

Other possible sources of sampling bias for effluents were analyzed in Phase I (ChemRisk 1994a; Ripple et al. 1996). That study concluded that the estimates of effluent releases were too low. The estimated bias factor developed for plutonium concentration estimates during the period 1953–1973 was lognormal, with a median of 1.3 and a geometric standard deviation of 1.6. That analysis did not address bias from non-representative sampling or from the choice of the alpha-particle self-absorption correction factor used at Rocky Flats.

Voillequé (1999b) evaluated the self-absorption correction factor that was used at Rocky Flats. Analysis of data of self-absorption from a variety of sources led to the conclusion that the value that had been used at Rocky Flats underestimated the plutonium activity on air sampling filters. The correction was estimated to be lognormal, with a median value of 1.16 and a geometric standard deviation of 1.05.

The overall estimate of bias considers all the factors discussed above. The distribution of estimates is approximately lognormally distributed with a median of 1.5 and a geometric standard deviation of 1.9. In the Monte Carlo calculations in Section 5, this estimate of bias is used together with the estimate of 170 g carried to the main filter bank (Table 3.4, Figure 3.3) to characterize the earlier deposition of $^{239/240}Pu$ on the filters.

3.2.2.2 Filter Contamination from Secondary Exhaust Feeder Ducts. Table 3.2 shows that only about half of the air transported to the inlet of the main filter bank was carried by ducts that were sampled. There were other ducts, with generally low flow rates, that were not sampled. These ducts exhausted ventilation air from a variety of locations in the building. Two of the ducts served the second floor, which did not contain any plutonium processing areas. However, the booster system filters and fans were located there, as was the main filter plenum. Ventilation

system components, steam boilers, chemical makeup areas, and electrical equipment were all located on the second floor.

Activities in some of the rooms whose ventilation exhausts were not sampled are of particular interest. These ventilation exhaust ducts carried some air from Rooms 179 and 180 in the development area. Plutonium fabrication, including casting and machining, was performed in gloveboxes in Rooms 181–182. The glovebox exhaust air from this area was handled by the recirculating air system (Table 3.3). Plutonium components were stored in Rooms 187–188. The reviews of Chew and Associates (1992) and ERFOM (1994) indicate that these are the areas most likely to have had airborne plutonium contamination.

The Building 71 room air sampling program provides information about possible contributions to the main filter bank loading from these sources. A program for sampling ambient air in rooms and hallways was initiated at the time the facility began operations. The room air samplers were designed to obtain estimates of airborne activity to which workers might be exposed and to identify locations where releases from gloveboxes or other systems were occurring. The measured concentrations do not necessarily reflect average air concentrations in the rooms, but they provide indications of the areas where substantial airborne contamination occurred and of the areas were airborne activity was found only infrequently. Special air samples were also collected to measure concentrations in work areas when it was known that there would be elevated air concentrations.

Initially, the samples were collected only during the day shift. Later, as the work schedule increased, two- and three-shift sampling was implemented. The room air samples were counted for total long-lived alpha activity and the Koval (1945) method was used to correct for naturally occurring radioactivity. Room air sampling data were reported using units of percent of maximum permissible level (MPL). At the time, the MPL for $^{239/240}$Pu was 4×10^{-12} µCi cm^{-3} (4pCi m^{-3} or 8.88 disintegrations per minute per cubic meter [dpm m^{-3}]). This is a very small mass of airborne plutonium, approximately 5.6×10^{-11} µg cm^{-3} (56 pg m^{-3}).

The results of measurements from this sampling program are tabulated in Appendix D. Before May 1955, plutonium air concentrations were overestimated because of inadequate correction for the presence of naturally occurring radionuclides. The change, resulting from an improved counting procedure, was most obvious in areas where air concentrations were routinely low.

The room air sampling data collected before the 1957 fire are summarized in Table 3.5. The rooms are grouped according to the average air concentrations that were measured during that period. Hallways are not listed individually, but nearly all of those results fall in the lowest category of 0–20% MPL (<0.8 pCi m^{-3}). Hallway 196 was the exception, with an average concentration of 23% MPL (<0.9 pCi m^{-3}). Examination of the data shows that most of the rooms the measured concentrations of $^{239/240}$Pu were highest (greater than 50% MPL or 2 pCi m^{-3}) were exhausted through ducts that were routinely sampled.

Table 3.5. Average Pre-fire Air Concentrations Measured in Rooms in Bldg. 71

Percentage of maximum permissible level (MPL)					
0–20[a]	20–50[b]		50–100[c]	100–200[d]	> 200[e]
Room 108	Room 125	Room 160	Room 149	Room 151	Room 146
Room 135	Room 129	Room 164	Room 180	Room 181	Room 148
Room 147	Room 141	Room 166	Room 186	Bldg. 74	Room 148A
Room 155	Room 152	Room 182			Room 149A
Room 179	Room 153	Room 235			Room 149B
Room 179A	Room 156	Room 247			Room 187
Room 180A	Room 158	Room 248			
Room 233	Room 159				
Room 240					
Room 283					

[a] Concentrations of $^{239/240}$Pu less than 0.8 pCi m^{-3}.
[b] Concentrations of $^{239/240}$Pu in the range 0.8–2 pCi m^{-3}.
[c] Concentrations of $^{239/240}$Pu in the range 2–4 pCi m^{-3}.
[d] Concentrations of $^{239/240}$Pu in the range 4–8 pCi m^{-3}.
[e] Concentrations of $^{239/240}$Pu greater than 8 pCi m^{-3}.

Some rooms with higher concentrations were not covered by the duct sampling program. In the highest category, Room 187, which was sampled for less than 1 year, showed consistently elevated concentrations, with an average of 337% MPL (13 pCi m^{-3}). It is not known whether the failure to sample reflected a lack of activity in the plutonium component storage room. The record does not exclude the possibility that elevated concentrations had occurred there for a number of years. Air concentrations in the other plutonium part storage room (Room 186) were measured over the entire period and were lower. The concentration there averaged 85% MPL (3.4 pCi m^{-3}).

Average concentrations of $^{239/240}$Pu in the fabrication area (Rooms 181 and 182) were also elevated. The higher levels in Room 181 were primarily due to high concentrations in the period October 1954–February 1955. The average for the pre-fire period was 117% MPL (4.7 pCi m^{-3}). Concentrations in Room 182 were lower, averaging 29% MPL (1.2 pCi m^{-3}).

Four rooms on the second floor of the building had pre-fire average $^{239/240}$Pu concentrations in the range 20–50% MPL (0.8–2 pCi m^{-3}). Reference flow rates for exhaust serving areas on the second floor totaled about 11,000 ft^3 min^{-1}. This exhaust flow amounts to about 453,000 m^3 d^{-1}. If air were discharged to the plenum continuously at the reference flow rate during the roughly 1530 days between startup of operations and the fire, the total volume of air would be 6.9×10^8 m^3. If the average concentration in all of the exhausted air were 2 pCi m^{-3}, the total amount of $^{239/240}$Pu carried to the plenum filters would be 1.4×10^9 pCi. This $^{239/240}$Pu activity corresponds to a plutonium mass of 1.9×10^{10} pg or ~0.02 g.

Similar calculations have been performed for the other ducts that were not sampled routinely. It is possible that the unmeasured air concentration in Room 187 was high throughout the period of interest. Perhaps the average concentration of $^{239/240}$Pu in the exhaust from Rooms 186–188 was 20 pCi m^{-3}. If the reference flow rate (2400 ft^3 min^{-1}) for the storage was maintained continuously, the total amount of $^{239/240}$Pu carried to the filter plenum would be about

0.04 g. The reference flow rate for the fabrication area is about 10 times higher (~26,000 $ft^3\,min^{-1}$ in all). Average concentrations measured in Rooms 181–182 were about 3 pCi m^{-3}. Using assumptions similar to those stated above, the estimated amount of $^{239/240}Pu$ carried to plenum filters from the fabrication area was ~0.07 g.

In summary, estimates based on the room air sampling data indicate that contributions to the $^{239/240}Pu$ loading on the main plenum filters did not exceed 1 g. This is a small fraction of the loading estimated for the ducts carrying air from the main process areas. The uncertainty band for that estimate is broad enough to include the minor contribution from unsampled ducts.

3.2.3 Plutonium Collected on the Booster System Filters

The fire in the booster system filters is believed to have rapidly consumed them (see Table 2.2. Plutonium that was made airborne during that fire was carried to the plenum filters and some of it penetrated through those filters. The methods used to estimate the amount of plutonium that had collected on the booster system filters are discussed below.

Inputs to the booster system came from the filtered glovebox exhaust ports in the analytical and development areas, including those from gloveboxes in Room 180. Concentrations of airborne plutonium were measured in the outlet from the booster system as part of the duct sampling program that was described above (Section 3.2.2.1). Reported measurement results are given in Appendix C. The reported concentrations have been used to make initial estimates of the $^{239/240}Pu$ activity that passed through the filters and was carried to the main filter bank. Figure 3.3 is a plot of the amounts of $^{239/240}Pu$ that passed through the booster system filters between plant startup and 11 September 1957.

The amount of plutonium on the booster filters at the time of the fire depends upon the history of booster system filter changes. One change of the booster system filters before the 1957 fire has been positively identified from review of plant reports and logs (Putzier et al. 1953–1958; Anonymous 1957). The time of the known filter change is indicated in Figure 3.4. The plot of the amount of activity leaving the booster system filters suggests that other changes of these filters may have occurred. However, documentation has not been found to verify that other filter changes occurred.

The initial estimate of the cumulative amount of $^{239/240}Pu$ that passed through the booster system filters was 530 µCi (Table 3.4). This quantity (7.4 mg) can be used to estimate the amount collected by the filters by making various assumptions about the efficiency of these filters. It has been assumed that there were no additional changes of the booster system filters. There were two sets of booster system filters through which the air passed sequentially. In the Monte Carlo analysis of releases, the efficiency of each set of booster system filters is considered to range between 98 and 99% (see Section 5). This exemplary calculation uses a central value of 98.5% for the efficiency of each set of filters. The following relationships apply for each set of filters.

$$I = \frac{\varepsilon E}{(1-\varepsilon)} \tag{3.1}$$

$$T = \varepsilon I \tag{3.2}$$

where
I = the amount of plutonium (g) carried to the filter inlet
ε = the collection efficiency of the filter
E = the amount of plutonium (g) that penetrated through the filter
T = the amount of plutonium (g) trapped by the filter.

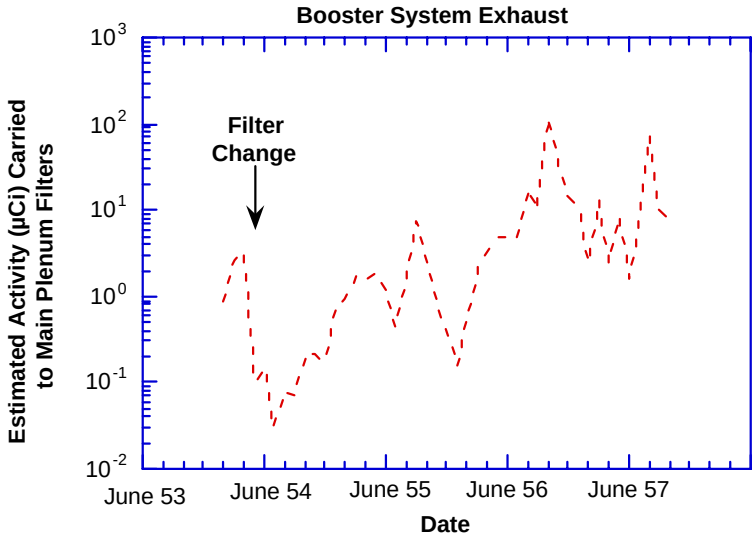

Figure 3.4. Initial estimates of the amounts of $^{239/240}$Pu that passed through the booster system filters and were carried to the main filter bank.

The initial estimate of the amount of E for the second filter set was 0.0074 g. The central value of ε was 0.985. The preliminary estimates of the amount of plutonium on the second and first filter sets were 0.48 g and 32 g, respectively. The preliminary estimate of the total amount of plutonium that was on the booster filters at the time of the fire was 33 g. A preliminary estimate of the average rate of accumulation of plutonium on the booster system filters was 0.021 g d^{-1}.

The need to make corrections to estimates based upon Appendix C was discussed in Section 3.2.2.1. Three types of corrections (for nonrepresentative sampling, for measurement bias, and for self-absorption of alpha particles) were identified there. Because the booster system exhaust line was only 18 in. in diameter and the sampling point was downstream of the booster system exhaust fan, samples collected from that line were very likely to be representative. No correction for non-representative sampling was applied to the booster system exhaust data. The measurement correction factor identified in Phase I (ChemRisk 1994a; Ripple et al. 1996) was characterized by a lognormal distribution with a median of 1.3 and a geometric standard deviation of 1.6. The correction for self-absorption of alpha particles (Voillequé 1999b) was also found to be lognormal, with a median of 1.16 and a geometric standard deviation of 1.05. The combination of these factors yielded a composite correction factor whose distribution was lognormal, with a median of 1.5 and a geometric standard deviation of 1.65.

In the Monte Carlo calculations of releases in Section 5, initial estimate of activity passing through the booster filters (7.4 mg) was modified using the two correction factors applicable to

the measurement procedures. The range of filter efficiencies considered there was 0.98–0.99; a uniform distribution of efficiencies over this range was assumed. This range is consistent with the observed performance of the main exhaust filters before the accident.

There has been some confusion about the nature of booster filter contamination in Building 71. The following discussion is presented to distinguish between the booster system present in 1957 and another system studied years later under different conditions.

After the 1957 fire, the primary mission of Building 71 was plutonium recovery. Many changes were made in the ventilation filtration systems to address operational problems. As noted earlier, the incinerator was a major source of plutonium contamination in the ventilation system in the early 1960s (Owen 1963). During 1962–1964, the capability of plutonium recovery operations in Building 71 was increased to about 600 kg month^{-1}. This was a major change from the original design throughput of about 25 kg month^{-1} (Chew and Associates 1992; ERFOM 1994). Figure 3.5 illustrates the increase in production that occurred in Building 71. The figure also charts the increase in the badged worker population at Rocky Flats as an indicator of the overall increase in production between 1953 and 1970. Both parameters are measures of the increase in Rocky Flats production operations over time.

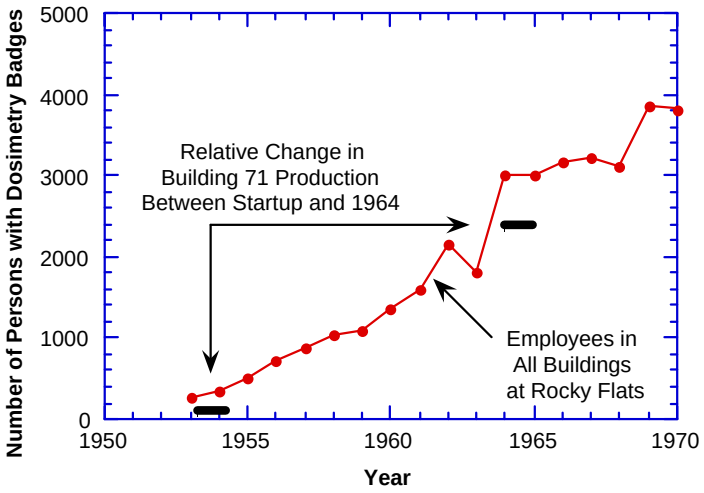

Figure 3.5. Changes in Rocky Flats operations with time after startup.

Several years after the large increase in processing rate, there was a study of plutonium buildup on filters in Booster System No. 3 (Woodward 1971). That system was installed to handle exhausts from gloveboxes in the expanded plutonium recovery processing line. The buildup of plutonium on the filters of Booster System No. 3 was estimated to average 13 g d^{-1}. There were large differences between the scope and nature of the development and analytical area operations during 1953–1957 and the massive plutonium recovery operations that occurred in Building 71 during the early 1970s. For that reason, the reported measurements of plutonium buildup on the filters of Booster System No. 3 (Woodward 1971) are not representative of the buildup of $^{239/240}$Pu on booster system filters prior to September 1957. The estimates made by Woodward for Booster System No. 3 are even less representative of the buildup of $^{239/240}$Pu on main plenum exhaust filters before September 1957. In Fairfield and Woods (1978), the data

from Woodward (1971) are used incorrectly to estimate the plutonium content of the main filter plenum before the fire.

3.2.4 Plutonium Collected on the Glovebox Outlet Filters and Room Air Prefilters

The booster system collected air from about 130 filtered exhausts from gloveboxes in the development and analytical areas of Building 71. Twenty-eight of these glovebox exhaust lines were located in Room 180 (Austin 1952). The total amount of plutonium that had been collected on glovebox outlet filters can be estimated using Equations (3.1) and (3.2), given above. An example calculation is presented here. The preliminary estimate of the total amount of plutonium that escaped from the glovebox outlet filters was about 33 g. The average efficiency of the glovebox outlet filters is estimated to range from 90 to 95%. The central value of 92.5% is used in this preliminary calculation. The approximate central estimate of the total amount of plutonium on all of the glovebox outlet filters is 400 g.

In the Monte Carlo calculations described in Section 5, estimates of the amount of plutonium on glovebox filters reflect the stated range of glovebox filter efficiencies. They also reflect the range of booster filter efficiencies given above and the measurement uncertainties associated with the amount of plutonium in the booster system exhaust.

The plutonium loading on individual filters is estimated to be about 3.1 g. In the Monte Carlo calculations, this quantity also depends upon the variables identified above. Photographs in the fire investigation report (Epp et al. 1957) were used to examine the extent of the fire and to estimate the number of filters burned. It is estimated that 16 glovebox filters in Room 180 were consumed in the fire.

Prefilters for the Room 180 ventilation air exhaust ducts were also burned in the fire. Estimates of the amount of plutonium that had been collected by those filters are also needed. Table 3.4 shows the preliminary estimate of the total amount of plutonium carried to the filter plenum by the D-88 exhaust duct. That duct collected air from all of the development and analytical areas, including Room 180.

The activity on the Room 180 prefilters can be estimated by assuming that all the plutonium carried by the D-88 exhaust duct originated in that room. Table 3.5 shows that the average concentrations in that room were highest among the rooms in the development and analytical areas. If the nominal efficiency of the prefilters were as high as that for the booster system filters (98.5%), this approximate calculation yields an estimated plutonium loading of 0.09 g on the prefilters. This estimate is inexact and does not consider the correction factors. Nonetheless, it can be concluded that the amount of plutonium on these prefilters was not an important source of plutonium in the 1957 fire.

3.3 Plutonium Deposited in the Ventilation System

Plutonium particles that are carried in air through a ventilation duct will deposit on the walls of the duct. When the particles are large or dense and when the air velocity is low the amount of deposition will be increased. Conditions conducive to plutonium deposition in exhaust lines occurred at Rocky Flats. During the period of early operations, there were two sets of lines where plutonium deposition was most likely. These were the glovebox exhaust lines in the recirculating air system, which served the main production gloveboxes, and those in the booster exhaust

system, which served the development and analytical areas. Earlier deposition of plutonium in the portion of the booster exhaust system that was involved in the fire is estimated in this section.

Buildup of plutonium deposits in glovebox exhaust system lines was addressed as an operational problem in the early 1970s (Cruickshank et al. 1974). About 375 ft of lines that exhausted gloveboxes in plutonium processing areas were replaced between 1972 and 1974. However, an inspection team found, in late 1974, that the total amount of plutonium in glovebox exhaust lines had not been estimated and that some glovebox exhausts were not filtered at the outlet (Cruickshank et al. 1974). The survey team also found that there was no regular schedule for changing glovebox outlet filters and few records of changes that had occurred. The absence of outlet filtration and intentionally induced failures (which have been reported by former workers) of filters that had been in operation so long that they were plugged would both lead to further releases into the glovebox exhaust lines and greater deposition of plutonium in those lines.

Following the inspection, a survey of glovebox exhaust lines in Building 71 was performed. At that time (1975), because of the expansion in recovery operations that was discussed in Section 3.2.3, there were many more glovebox exhaust systems in operation. There were eight booster systems for processing gloveboxes and one for the research and development gloveboxes. Nearly all of the 17.6 kg of plutonium that was estimated to be in glovebox lines was associated with the main recovery operations. More than 3 kg of plutonium had been removed from one section of a processing glovebox line. Another 470 ft of processing glovebox line, estimated to contain nearly 8 kg, was being replaced. In contrast, the total amount of plutonium in the 750 ft of research and development glovebox lines was estimated to be 130 g (Bowman 1975).

None of the lines in the development area were among those that had been replaced during the 1972–1974 period identified above. It is not known whether the glovebox outlet lines in Room 180 were removed or whether the exterior surfaces were simply decontaminated during the cleanup of that room in 1960–1961. There were some further modifications of the development area and of Room 180 in 1965, but it is not known whether those changes significantly affected the glovebox outlet lines.

It is known that glovebox outlet filters were in use in the development area before the 1957 fire. This would tend to minimize the amount of plutonium in the lines and is consistent with the finding, nearly 20 years later, of small quantities of plutonium in those lines. If the rate of accumulation of plutonium in the lines was constant over the period, it is estimated to be about 6 g y^{-1}. In the approximately 4-year period between plant startup and the fire, accumulation of plutonium in the booster system is estimated to be about 25 g. An uncertainty range of a factor of 2 is assumed in the calculations described in Section 5.

A comparable amount (25 g) of plutonium is assumed to have been attached to Plexiglas structural components. This is expected to have occurred primarily as the result of spills onto the floors of the gloveboxes.

3.4 Summary of Amounts of Plutonium Involved in the Fire

The estimated amounts of plutonium that were present in Room 180 and elsewhere in the path of the fire are summarized in Table 3.6. The quantities shown are those estimated to be present at the time the fire began. The uncertainty estimates for these quantities are also included in the table. For some quantities, such as the amount of plutonium in lathe oil and sludge and the

amount of plutonium metal, the uncertainty is reflected as a range of possible amounts. The values given for the amounts of plutonium on filters are the preliminary nominal estimates. A range of values was computed, as described in the corresponding footnotes. The range reflects the measurement uncertainties for quantities used to derive the nominal estimates (Section 3.2) as well as uncertainties in filtration efficiencies.

Table 3.6. Estimated Amounts of Plutonium Involved in the Fire

Location and form	Nominal amount (kg)	Uncertainty distribution type and parameter values
Room 180		
Pu in organic liquids	1.7	Uniform, 1.0–2.4 kg
Pu metal pieces and components	12	Uniform, 9–16 kg
Pu dioxide in can	2.6	Uniform, 2.5–2.7 kg
Pu collected in glovebox outlet filters that burned	0.050[a]	[b]
Pu contamination of Plexiglas glovebox walls	0.025	Uniform, 12–50 g
Pu deposited in glovebox exhaust lines	0.025	Uniform, 12–50 g
Filter systems		
Pu collected on booster system filters	0.033	[c]
Pu collected on main plenum filters	0.17	Lognormal, GM = 1.5, GSD = 1.9[d]

[a] This is the nominal estimate for the 16 glovebox outlet filters in Room 180 that burned during the fire.

[b] The quantity of plutonium on the glovebox outlet filters is computed as part of the Monte Carlo calculations. The range of values depends upon the calculations for the booster system filters (note c) and the uncertainty in the outlet filter efficiency (uniform, 0.90–0.95).

[c] The quantity of plutonium on the booster system filters is computed as part of the Monte Carlo calculations. The range of values depends upon the uncertainties in the measurements of the booster exhaust and upon the uncertainty in the efficiency of the booster system filters. The measurement uncertainty is characterized by a lognormal distribution with GM = 1.5 and GSD = 1.65.[d] The efficiency of the booster system filters is characterized by a uniform distribution over the range 0.98–0.99.

[d] GM is the geometric mean of the lognormal distribution and GSD is the geometric standard deviation of the lognormal distribution.

4. FRACTIONS OF PLUTONIUM RELEASED INTO AIR DURING FIRES

Section 3 described the various sources of plutonium that were involved in the 1957 fire and were potentially available for release to the air during the fire. The next step (see Figure 1.2) is to estimate, for each source, the fraction of the plutonium that was suspended into the air as a result of the fire. The airborne plutonium was then transported by the booster system and room ventilation systems to filters or, when those had failed, to the stack and the environment. This section describes the release fractions for plutonium in various forms under conditions that are either known to have occurred or may have occurred during the fire.

The fractional release of plutonium into the air depends upon the chemical and physical form of the material and the forces that are applied. Estimates of airborne release fractions for various situations are based upon experimental studies. These tests employed plutonium that was heated, oxidized, or subjected to other stresses under controlled conditions.

Several different chemical forms were present in the area affected by the fire (Section 3.1). Most of the plutonium was present as the metal, mainly in the form of hemispheres. Metal turnings were also present in lathe oil and sludge. Plutonium dioxide (PuO_2) powder was stored in a can. Plutonium had also collected on filters in various locations (Section 3.2). This plutonium had been continuously exposed to air and would be expected to be present as the oxide. Plutonium was also deposited in the booster system exhaust lines that led from the glovebox outlets to the booster system filters. These deposits would also be expected to be plutonium oxide because of their previous exposure to air flowing through the booster system.

A review of measurements of releases of plutonium and plutonium simulants in various types of fires was commissioned to support emergency planning activities by the State of Colorado and the Rocky Flats Plant. The report (Kogan and Schumacher 1993) contains recommendations for estimating airborne release fractions and respirable fractions of aerosols released from a variety of fires that may occur in the future. Kogan and Schumacher (1993) reviewed original reports of plutonium release measurements, examined other reviews and recommendations, and prepared their own recommendations for use at Rocky Flats. Their report provides recommended release fractions for three categories of interest: oxidation of plutonium metal, burning of organic liquids containing plutonium metal, and burning of solids contaminated by plutonium.

More recently, a standard has been developed to provide a uniform approach for selecting airborne release fractions used in accident consequence assessments (ANS 1998). That document addresses accidental releases at nonreactor nuclear facilities. Appendix A of the ANS standard provides a summary of bounding airborne release fractions that have been peer reviewed and whose basis is supported by data from one or more experimental investigations. That appendix is based primarily upon information analyzed and compiled in a comprehensive report on this topic (DOE 1994b, 1994c). These documents contain an extensive review of experimental data relevant to assessing release fractions for a variety of accidents.

The U.S. Nuclear Regulatory Commission (NRC) has also published a new handbook for analyzing the effects of accidents in nuclear fuel cycle facilities (SAIC 1998). The NRC handbook also recommends airborne release fractions for various accident situations. Many of the recommendations in the NRC handbook are based on information and evaluations presented in the earlier U.S. Department of Energy (DOE) handbook (DOE 1994b, 1994c). A large fraction of the recommended values in the NRC handbook are included in Appendix A of the ANS standard (ANS 1998).

Most accident analyses are prospective assessments of events that are considered unlikely but possible. The term "maximum credible accident" has been used to describe such an event. In general, the goal of such an analysis is to provide bounding estimates (upper limits) for the consequences of the accident being evaluated. This goal often leads to very cautious (also called "conservative") assumptions regarding various parameters used in the calculations. The present retrospective analysis of an actual event attempts to make realistic estimates of the releases. The range and distribution of release estimates are calculated using the Monte Carlo procedure described in Section 5 and reflects uncertainties in all parts of the calculations.

The following sections discuss information about airborne release fractions for various situations that are relevant to the 1957 fire. Section 4.1 addresses oxidation of plutonium metal. Sections 4.2 and 4.3 deal with combustion of plutonium contaminated liquids and solids, respectively. Section 4.4 considers the release fraction for plutonium dioxide powder. Section 4.5 addresses the question of resuspension of previously deposited plutonium. Release fractions for filters subject to blast effects are described in Section 4.6. All of the release fractions and the associated uncertainty distributions are summarized in Section 4.7.

4.1 Airborne Release Fractions for Plutonium Metal Oxidation

Airborne release fractions for oxidation of plutonium metal depend upon several factors. The metallurgical type and temperature of the metal are quite important. Under some conditions, the surface area of the metal, the humidity of the air, and the air velocity have also been shown to be important considerations when estimating release fractions.

4.1.1 Oxidation at Temperatures Below the Melting Point of Plutonium

Kogan and Schumacher (1993) recommended methods for calculating airborne releases of plutonium oxidized at temperatures below the melting point. They considered both alpha- and delta-phase plutonium. Their recommendations for alpha-phase plutonium, the type present in Room 180, are discussed here. Alpha-phase plutonium has a density of 19.8 g cm^{-3} and a melting point of 640°C (1180°F). For static conditions, the airborne release rate (*ARR*, µCi cm^{-2} h^{-1}) depends upon the absolute (Kelvin scale) temperature (T_a, K) and the absolute humidity (*AH*, mg L^{-1}). The relationship is given by Equation (4.1), which relates the logarithm of the airborne release rate to those variables. Kogan and Schumacher (1993) presents information that indicates that minimum and maximum values of the *ARR* measured under the same conditions differ by factors of 2–5.

$$\log (ARR) = (-4.36 + 0.071\ AH)\ (0.23 - 350.6\ T_a^{-1} + 1.91 \times 10^5\ T_a^{-2}) \quad (4.1)$$

It is necessary to estimate the area of the metal hemispheres in order to use Equation (4.1) to predict the total amount of plutonium made airborne. The dimensions and masses of the hemispheres that were present in the glovebox line are not described in available documents. Public statements have been made to the effect that the critical part of the plutonium bomb used at Nagasaki was "the size of a billiard ball" (true billiards, pool, or snooker not specified). That uncertain information about the diameter, together with the upper bound estimate of 1.8 kg for the average mass of the hemispherical shells, has been used to estimate a range of surface areas of 150–300 cm^2.

The airborne release rate from a hemisphere that was oxidized was estimated for an absolute humidity of 16 mg L^{-1} and a temperature of 500°C (930°F). That temperature (773 K) is near the ignition temperature of a plutonium component that has a low surface to mass ratio. For those conditions, the *ARR* is about 0.5 µCi cm^{-2} h^{-1} or about 7 µg cm^{-2} h^{-1}. Considering the range of estimated areas of hemispheres involved in the fire and the fire duration of about 40 minutes, the total release is estimated to be between 0.7 and 1.4 mg of plutonium. The fraction of the original mass (1800 g) that would be released is estimated to be in the range 4–8 x 10^{-7}.

For a lower temperature of 350°C (523°F), the computed *ARR* is about 0.18 µCi cm^{-2} h^{-1} or about 2.5 µg cm^{-2} h^{-1}. A central estimate of the *ARF* for that temperature is about 2 x 10^{-7}. The bounding value given in Appendix A of the ANS standard for plutonium oxidation at less than the ignition temperature is 3 x 10^{-5} (ANS 1998). That is the same bounding estimate that is given in the DOE and NRC handbooks (DOE 1994b, 1994c; SAIC 1998). It is much higher than the estimate made using Equation (4.1), but it does not explicitly consider the surface area involved or the time period. Data compiled in the DOE (1994b, 1994c) handbook for these conditions show an experimental *ARF* for alpha-phase plutonium of 1.3 x 10^{-6}, which is about 8 times higher that the estimate from Equation (4.1). The bounding value and most of the experimental data considered were for delta- and beta-phase plutonium.

In general, processes produce or suspend a range of particle sizes. For plutonium that is released to the environment and may be inhaled, it is the respirable fraction (*RF*) of the aerosol that is of interest. As a general rule, particles with aerodynamic diameters exceeding 10 µm do not penetrate into the pulmonary region of the lung (ICRP 1994; Grogan et al. 1999). For that reason, the respirable fraction consists of the particles having aerodynamic diameters that are less than 10 µm. For PuO_2 particles, which have a density of 11.5 g cm^{-3}, respirable particles are those having physical diameters less than 3 µm.

Kogan and Schumacher (1993) recommended an equation for calculation of the respirable fraction of the aerosol produced under these conditions. The respirable fraction was considered to be a function of temperature, measured on the Celsius scale (T_c, C) as shown in Equation (4.2).

$$RF = 1.07 - 0.00353\ T_c + 3.82 \times 10^{-6}\ T_c^2 \qquad (4.2)$$

Kogan and Schumacher (1993) show measured respirable fractions for alpha-phase plutonium, by two different investigators, ranged from 0.9–1 at 22°C (72°F) to 0.6–0.7 at 123°C (253°F). One measured value for alpha-phase plutonium at 350°C was 0.2. Equation (4.2) bounds the measurement data that they compiled from the two studies. The computed value from Equation (4.2) for a temperature of 500°C is 0.26. For a temperature of 350°C (523°F), Equation (4.2) yields an estimate of the respirable fraction of 0.30, which is 50% greater than the measured value for alpha-phase plutonium oxidized at 350°C. It was, however, consistent with a measured *RF* for delta-phase plutonium at that temperature.

The generic respirable fraction listed in Appendix A of the ANS standard is 0.04 (ANS 1998). That value is based upon a particle size distribution measured for oxidation of delta-phase plutonium at 123°C (DOE 1994b, 1994c).

4.1.2 Oxidation at Temperatures Above the Melting Point of Plutonium

For combustion of plutonium at temperatures between the melting point and boiling point of 3235°C (5855°F) of plutonium, Kogan and Schumacher (1993) did not provide any relationship of the release fraction data with temperature. For static conditions, the authors recommended that respirable release fraction be estimated using the product of an *ARF* of 2×10^{-4} as and a respirable fraction (*RF*) of 0.5. This product of 1×10^{-4} was also recommended, for different reasons, for dynamic conditions in this temperature range. Variability among experimental measurements suggests that the uncertainty range for the recommended value could be as high as an order of magnitude.

Appendix A of the ANS standard (ANS 1998) gives a bounding value of the *ARF* for plutonium oxidation above the ignition temperature of 5×10^{-4}. The respirable fraction estimated for the same conditions is 0.5. The product, *ARF* x *RF*, for that condition is 2.5 times greater than that recommended by Kogan and Schumacher (1993).

The data tabulated in the DOE (1994b, 1994c) handbook for oxidation above the ignition temperature include four results for alpha-phase plutonium. Measured airborne release fractions ranged from 3.9×10^{-6} to 2.4×10^{-3}. The mean of 4.6×10^{-4} is strongly influenced by the one very large value. Three of the measurements were made using large pieces (0.45–1.7 kg). For those tests, the mean *ARF* was 2.1×10^{-4}. Measurements of particle size were reported for two of the tests. Mass median diameters of 8 and 11 µm were found. These results support selection of a *RF* ~0.5.

For a free fall spill of molten plutonium, Appendix A of the ANS standard (ANS 1998) recommends a bounding value of 0.01 for the *ARF*. The recommended *RF* for this condition was not given in Appendix A (ANS 1998), but the DOE (1994b, 1994c) handbook gives a value of 1 based on experiments in which plutonium droplets fell 0.75 m (2.5 ft).

For other dynamic conditions at elevated temperatures, release fractions estimated by Ballerau (1991) are of possible interest. Kogan and Schumacher (1993) cite his recommendations for conditions of high air velocity and for "vigorous" oxidation of plutonium. For air velocities in excess of 1.5 m s^{-1} and temperatures in the range 800–1000°C (1470–1830°F), an *ARF* of 5×10^{-4} and a *RF* of 1 were recommended. The "vigorous" oxidation conditions are even more severe, including projection of droplets and sparks, with temperatures in the range 1500–2000°C (2700–3600°F). For this condition, Ballerau recommended an airborne release fraction of 0.1 and a respirable fraction of 0.01.

No conditions that are directly comparable to those just listed are addressed in Appendix A of the ANS standard (ANS 1998) or the underlying documentation (DOE 1994b, 1994c). A violent dispersal of molten plutonium in which large drops (diameters of hundreds of micrometers) travel for distances of more than 1 m is addressed there. In this case, the explosive force comes from within the metal itself rather than from an external source. Examples of this type of event are exploding wire experiments in which a large electrical discharge is applied to destroy the wire. These results are definitely not applicable to events during the 1957 fire.

4.2 Airborne Release Fractions for Plutonium in Organic Liquids That Burn

For burning of contaminated organic liquids, Kogan and Schumacher (1993) recommended two values for the *ARF*: 0.1 for a dissolved contaminant in a tributyl phosphate (TBP) and kerosene mixture and 0.02 for powdered plutonium in kerosene. A respirable fraction of 1 is

recommended in both cases unless the respirable fraction of the powder was known. The value of 0.1 is nearly an upper bound for all experimental data; some measured release fractions are much lower (10^{-3}–10^{-2}). Neither of the conditions is exactly the same as the situation, at the time of the fire, in which particles of plutonium were present in cutting oil or sludge.

Appendix A of the standard (ANS 1998) gives a bounding value of 0.1 for vigorous burning, with surface turbulence, of the organic liquid to complete dryness. This is an upper bound for a broad range of values found experimentally (DOE 1994b, 1994c). The experiments employed a mixture of TBP and kerosene. When burning does not continue to complete dryness, a bounding *ARF* of 0.03 is recommended (DOE 1994b, 1994c; ANS 1998). In both cases, the *RF* was taken to be 1. The data on particle size were available from only one experiment (DOE 1994b, 1994c).

It has been suggested that the carbon tetrachloride present in the fire area could lead to production of plutonium chloride that would have a high release fraction. No experimental data specific to carbon tetrachloride were located. Because the amount of plutonium in the bottle of carbon tetrachloride is estimated to be quite small, the broad range of release fractions used is considered adequate to address this possibility.

4.3 Airborne Release Fractions for Burning of Plutonium Contaminated Solids

Kogan and Schumacher (1993) recommended two different values for burning of polymethylmethacrylate (PMMA, the main constituent of Plexiglas) with surficial contamination. If the contaminant was a solution or a salt, an *ARF* of 0.02 was recommended. If the contaminant was a dry powder, the value selected was 0.05. Experimental data show that *ARF*s from combustion of contaminated plastics are definitely higher those produced when cellulosic material is burned. The release apparently occurs primarily while the plastic is softening and material is out-gassing before flaming.

The DOE (1994b, 1994c) handbook also shows that *ARF*s for burning PMMA exceed those from burning cellulose. A bounding value for the *ARF* for contaminants on burning PMMA of 0.05 is estimated (DOE 1994b, 1994c). The *RF* for such releases is estimated to be 1.

For burning of cellulose and mixed solids contaminated with plutonium, Kogan and Schumacher (1993) recommended relationships that reflect a dependence upon the air velocity (u, m s^{-1}) through the fire. For the situation most like the combustion of filters, the particulate contaminants can be considered to be powders. For those conditions, Equation (4.3) was recommended for calculating the *ARF*. An upper bound for the *ARF* is 0.5 based on data for large fires.

$$ARF = 0.2754 \times 3.254^{\ln u} \tag{4.3}$$

Using a model of cellulose combustion and relevant data, Kogan and Schumacher (1993) estimated that, for accidental fires (but not jet fuel conflagrations) an appropriate value of u would be 8.3×10^{-3} m s^{-1}. When substituted into Equation (4.2), this estimate of u leads to a computed *ARF* of 9.7×10^{-4}, which is within a factor of 2 of release fractions measured for combustion of cardboard boxes of wastes. Kogan and Schumacher (1993) recommended an *ARF* of 1×10^{-3} for that type of release. The *RF* recommended for this type of release is one.

The DOE handbook gives a comparable upper bound for the airborne release fraction of 0.4. The respirable fraction of 1 is a cautious value chosen in the absence of relevant data (DOE 1994b, 1994c).

The airborne release factors for burning filters were estimated using Equation (4.3). For the booster system filters, the linear velocity of air through the filters was about 300 ft min^{-1} or about 1.5 m s^{-1}. For that velocity, Equation (4.3) predicts an *ARF* of 0.45, near the upper bound of values observed. It is also midway between the two recommended bounding values of 0.4 and 0.5. For the glovebox outlet filters, a range of air velocity from 0.8 to 1 m s^{-1} was identified. The range of *ARF*s for those filters is 0.22–0.28.

The air flow through the main filter plenum was low when the fire started. However, most of the filters in the main plenum were burned when the fans were on high speed or after the fans failed and natural draft conditions prevailed. From 10:25–10:40 p.m., the fans were operating at high speed. The air flow rate through each filter was about 480 ft^3 min^{-1}. This corresponds to a linear velocity of 2 ft s^{-1} or 0.61 m s^{-1}. For that air velocity, Equation (4.3) yields an *ARF* of 0.15 for the plenum filters. For later periods, many filters had burned through and it is not certain that air was flowing through each filter. However, a range of *ARF*s from 0.005–0.01 was estimated using Equation (4.3) under the assumption that there was flow through the burning filters.

4.4 Airborne Release Fractions for Plutonium Oxide Powder

Appendix B of the ANS standard (ANS 1998) provides some example calculations. For a fire that involved plutonium oxide in cans that did not have airtight lids, a bounding *ARF* of 6×10^{-3} was selected. This was based upon measurements of heating plutonium oxide in a stream of flowing air. The results were dependent on the air velocity, and the range of values for the highest velocity (1.2 m s^{-1}) ranged from 2.5×10^{-4} to 5.6×10^{-3} (DOE 1994b, 1994c). The *RF* for powdered plutonium oxide was taken to be 0.01 (ANS 1998).

4.5 Resuspension of Plutonium Deposited in Glovebox Exhaust Lines

Deposits of plutonium in the glovebox exhaust lines are estimated to have occurred during the first few years of operation before the fire. Deposited material could be resuspended during the course of the fire. Appendix A of the ANS standard (ANS 1998) provides a bounding estimate of the airborne release rate (*ARR*) for powder on surfaces, including steel, with an air velocity of less than 2 m s^{-1}. This velocity condition is consistent with the flow rate through the larger diameter piping where the deposits would most likely occur. The bounding estimate for the *ARR* is 4×10^{-5} h^{-1}. The corresponding *ARF* for suspension of the deposited plutonium in a 40-minute period would be about 3×10^{-5}.

4.6 Airborne Release Fractions for Plutonium on HEPA Filters Subjected to Blast Effects

Bounding estimates of the airborne release fraction for HEPA filters subject to blast effects are given in Appendix A of the ANS standard (ANS 1998). The estimated upper bound for the *ARF* from the pressure pulse from a deflagration is 1×10^{-2}. The bounding estimates were derived from experimental data on glass fiber HEPA filters. The bounding values were based

upon pressure differentials for simulated tornado conditions. The *RF* for such an event is assumed, in the absence of data, to be 1 (DOE 1994b, 1994c).

4.7 Summary of Airborne Release Fractions for Plutonium Involved in the Fire

Table 4.1 contains a summary of the airborne release fractions used in estimating plutonium release to the atmosphere during the 1957 fire. Uncertainties associated with the airborne release fractions are also included. When a central value was identified, an uncertainty distribution that covers a relatively narrow range was used. This was done to assure that the range of values employed in the Monte Carlo calculations will be consistent with the experimental basis used to estimate a particular *ARF*. In some cases, a range of values was identified and the central value was chosen to be the mean of the distribution. The bases for individual values are described in the footnotes to the table.

Table 4.1. Estimated Release Fractions for Plutonium Involved in the 1957 Fire

Location and form	Airborne release fraction (ARF)	Uncertainty distribution type and parameter values
	Room 180	
Pu in burning organic liquids	0.06 [a]	Uniform, 0.02–0.1 [b]
Pu metal oxidation category		
T < melting point (640°C)	1.3×10^{-6} [c]	0.6–2 x 10^{-6} [d]
T > melting point	2×10^{-4} [e]	Loguniform, 0.02–2 x 10^{-3} [f]
Dynamic, u > 1.5 m s^{-1}	5×10^{-4} [g]	Uniform, 0.25–0.75 x 10^{-3}
Vigorous, T > 1500°C	0.1 [g]	Uniform, 0.05–0.15
Pu dioxide in can	3×10^{-3} [a]	Uniform, 0.3–6 x 10^{-3} [h]
Pu collected in glovebox outlet filters that burned	0.28 [i]	Uniform, 0.24–0.32
Pu contamination of Plexiglas glovebox walls	0.035 [a]	Uniform, 0.02–0.05 [b]
Pu deposited in glovebox exhaust lines	3×10^{-5} [j]	Uniform, 1–5 x 10^{-5}
Pu collected by room air duct prefilters that burned	0.45 [k]	Uniform, 0.4–0.5 [b]
	Filter systems	
Pu collected on booster system filters	0.45 [k]	Uniform, 0.4–0.5 [b]
Pu collected on main plenum filters, burning	0.15 [l] 0.008 [a]	0.1–0.2 0.005–0.01 [m]
Pu collected on main plenum filters, deflagration	0.01 [j]	Uniform, 0.05–0.015

[a] Mean of distribution.

[b] Range of recommended bounding values.

[c] Value for alpha-phase plutonium.

[d] Lower bound computed using Equation (4.1); upper bound chosen to give a mean for the distribution that is equal to the observed value for alpha-phase plutonium.

[e] Mean of results for large pieces of plutonium.

[f] Broad range reflects wide range of observed values for alpha-phase plutonium.

[g] Value recommended by Ballerau (1991).

[h] Range of observed values.

[i] Derived using Equation (4.3) for u = 1 m s^{-1}.

[j] Recommended bounding value.

[k] Derived using Equation (4.3) for u = 1.5 m s^{-1}.

[l] Derived using Equation (4.3) for u = 1.0 m s^{-1} for fan operation at high speed during 10:25–10:40 p.m.

[m] Estimated range derived using Equation (4.3) for natural draft flow through burning filters after fans ceased operation.

5. PLUTONIUM RELEASE CALCULATIONS

Estimates of the amount of plutonium that was released to the environment during the fire depend upon many factors. The locations and amounts of plutonium involved in the fire were described in Section 3 and were summarized in Table 3.6. Information on release fractions to the air for plutonium in various forms and under various conditions has been discussed in Section 4. Airborne release fractions for material identified in Table 3.6 were presented in Table 4.1. The supplemented accident chronology was described in Section 2. That sequence of events, particularly the progress of the fire through the filter systems, is a primary factor affecting the time history of releases and also the magnitudes of releases at specific times.

In this section, we describe how information from Sections 2–4 is used in calculations of the plutonium release. As mentioned previously, the release estimates were obtained using Monte Carlo techniques to propagate the uncertainties through the steps of the calculations.

Table 2.2 showed the augmented chronology for the fire that was developed in Diliberto (1999). It indicates that the fire moved rapidly from the glovebox line to the booster system filters and then to the main filter bank. Glovebox outlet filters were nearest to the origin of the fire and were the first to be burned. It is estimated that the first of those filters was burned through by 10:07 p.m. The other glovebox filters that burned are estimated to have burned at the rate of about 1 per minute during the following 15 minutes. Plutonium that became airborne as the result of burning of these filters was carried through the booster system exhaust line to the booster system filters. While these filters were functioning, much of that airborne plutonium was trapped on those filters. However, as those filters burned through, their filtration efficiency was being reduced and eventually lost completely.

Estimates of the effectiveness of the booster and main plenum filters were developed using the chronology in Table 2.2. Figures 5.1 and 5.2 show the behavior of these filters as a function of time after the start of the fire. In the figures, filter performance is measured by the fractional penetration of plutonium particles through the filters. The fractional penetration (p) is the complement of the filter collection efficiency (ε) defined in Section 3.2.3. That is, $p = 1 - \varepsilon$. In Figure 5.1, both the central value and the uncertainty bounds for p are shown. The uncertainties in p were estimated using a uniform distribution centered on the best estimate. Based upon prior performance of the plenum filters, the pre-fire penetration through those filters was estimated to be 0.015, with a range of 0.01–0.02. The same initial value and range were used for each set of booster filters. However, those filters were burning soon after the start of the fire and failed completely within a few minutes.

Filters in the main plenum did not catch fire until 10:20 p.m. Initially, the fire progressed slowly because the ventilation system fans were operating at low speed with a flow of about 100,000 $ft^3\ min^{-1}$. As the plenum filters began to burn their performance was degraded. After some filters had burned through, much of the building exhaust air could pass through with little or no filtration. At sonic velocity, which is about 1870 $ft\ s^{-1}$ at an air temperature of 1000°F, an opening of only ~3 ft^2 is sufficient to handle a flow rate of 300,000 $ft^3\ min^{-1}$.

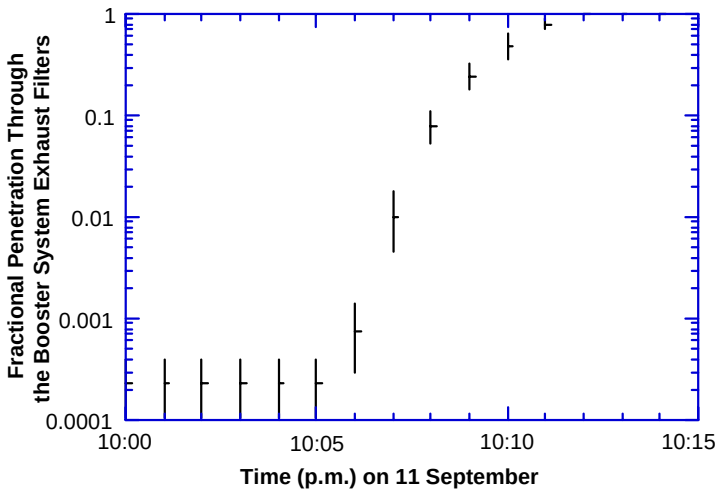

Figure 5.1. Estimated fractional penetration of particles through booster system exhaust filters. Central estimate and uncertainty bounds shown for times before filter failure. These filters became completely ineffective at about 10:12 p.m.

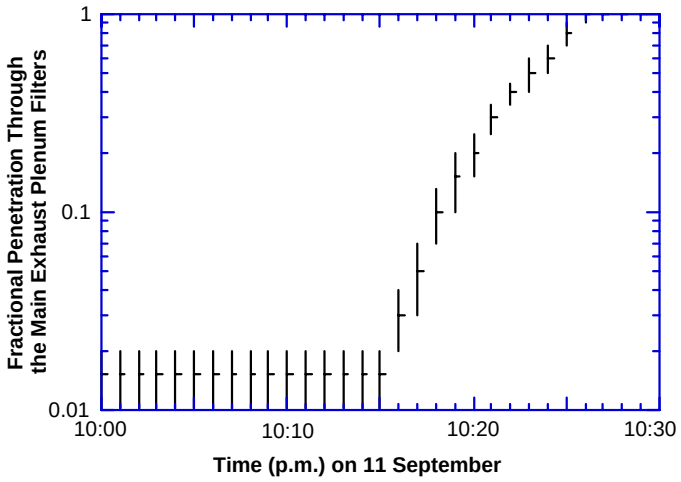

Figure 5.2. Fractional penetration of particles through the filters in the main exhaust plenum of Building 71. Central estimate and uncertainty bounds shown for times before filter failure. These filters became completely ineffective at about 10:26 p.m.

When the fire in Room 180 was discovered, flames were already outside the Plexiglas gloveboxes. By that time (10:10 p.m.), the normal airflow pattern from the room into the gloveboxes was disrupted. Releases of plutonium from the metal being oxidized and from the lathe oil and sludge being burned were then carried primarily into the air in Room 180. The booster system exhaust flow rate from gloveboxes affected by the fire was only about 640 $ft^3\,min^{-1}$. As the Plexilas glovebox components burned, the booster system connections to the gloveboxes were lost.

The room ventilation air passed through prefilters that were located in the exhaust ducts. Those prefilters were estimated to have caught fire at 10:17 p.m. and to have burned through quickly. Particle penetration through these filters is estimated to have increased rapidly beginning at 10:14 p.m. as their temperature increased. It is estimated that $p = 1$ for the prefilters at 10:19 p.m. Uncertainties in the estimates of the efficiencies of the prefilters were comparable to those estimated for a single bank of booster filters and were somewhat smaller than those indicated in Figure 5.1. After the time that the prefilters had burned through, there were no intact filters between the fire in Room 180 and the main filter plenum.

Figure 5.2 shows the estimated increase of particle penetration through the main filter bank as a function of time. The main filter bank is estimated to have failed before 10:30 p.m. After that time, airborne plutonium carried by ducts to the exhaust plenum was transported to the stack and to the environment without filtration.

Table 4.1 identifies four categories of release fractions for plutonium metal oxidation under various conditions. The actual conditions of plutonium oxidation during the fire are not known. For baseline estimates of plutonium releases, equal amounts of the plutonium metal inventory (9–16 kg [Table 3.6]) were assigned to each of the four metal oxidation categories. Table 5.1 illustrates the amounts of material in each category and bounding estimates of releases for the largest estimated amounts of plutonium that became airborne in Room 180. It is clear that assignment of equal amounts to the four metal oxidation categories is recognized to be an important scenario uncertainty that affects the estimated amounts released. Therefore, some alternative assumptions were considered to examine this factor and are discussed in Section 6.

Table 5.1. Largest Estimated Amounts of Plutonium That Became Airborne in Room 180

Location and form	Quantity of material in Room 180	Range of estimated airborne amount[a]
Pu in burning organic liquids	1.0–2.4 kg	20–240 g
Pu metal oxidation category	Total of 9–16 kg	
T < melting point (640°C)	25% of total	1.4–8.0 mg
T > melting point	25% of total	0.045–8.0 g
Dynamic, $u > 1.5$ m s^{-1}	25% of total	0.56–1.7 g
Vigorous, T > 1500°C	25% of total	110–340 g
Pu dioxide in can	2.5–2.7 kg	0.75–16 g

[a] Two significant figures shown only to illustrate the arithmetic results.

Rates of release are assumed to be linear during the period to which they are applicable. For the plutonium metal components that were in the gloveboxes, metal turnings that were in oil and sludge, and the can of plutonium oxide, the period of release is the duration of the fire in Room 180. The same period of release was used for plutonium contamination on the glovebox walls. For plutonium that had been collected by various filters, the release period was the time during which the filters were burning. Suspension of plutonium previously deposited in glovebox exhaust lines was assumed to continue throughout the time while the power was on and the booster exhaust system was operating.

It is indicated in Tables 3.6 and 4.1 and in the foregoing discussion that both the amounts of material and the applicable release fractions are uncertain quantities. Table 5.1 illustrates the bounds of some of the multiplication of the two parameters. The Monte Carlo calculation procedure uses information about the entire distributions of possible values for these and other parameters to make a series of estimates of the plutonium release amounts for various time periods during the fire.

The calculations entail the following steps. The release pathways to the atmosphere were identified (Figure 1.2). Central estimates and distributions of values for the quantities of plutonium involved in the fire were developed (Table 3.6). Distributions of estimates of release fractions, as well as central estimates, were developed for each of the plutonium amounts identified in Table 3.6. Those estimates were summarized in Table 4.1. Transport of the material that became airborne to the environment depended upon filter efficiencies. The most important of those are illustrated in Figures 5.1 and 5.2. In the calculations, each of the relevant distributions was sampled and the selected values were used to compute an estimate of the release for the time period under consideration. The calculations were repeated many times to produce a large number of estimates of the release quantity for that time period. The many results form a distribution of estimates of the particular release quantity that reflects the uncertainties of parameters used in its calculation.

Release estimates for the early part of the fire (10:00–10:45 p.m.) were computed at 1-minute intervals. For the next half-hour (10:45–11:15 p.m.), when conditions were not changing as rapidly, 5-minute average release estimates were calculated. Later still (11:15 p.m.–2 a.m.), the release estimates were made for 15-minute periods.

The primary results from the calculations are the releases during 15-minute periods. That duration corresponds to the time resolution of the meteorological data that are available for the time of the fire. Distributions of 15 one-minute estimates were summed to obtain the distributions of release estimates for each of the first three release periods. Distributions of three 5-minute release estimates were summed to obtain the distribution of release estimates for the next two release periods. No summation was needed for the time periods after 11:15 p.m. The estimated 15-minute releases are considered more reliable than estimates made for shorter time intervals because the latter are more subject to uncertainties in the fire chronology.

Results of the calculations and their uncertainties are discussed in the following section. In addition to the main results (15-minute release totals), the 1-minute release estimates are shown to illustrate how the release estimates changed with time during the early part of the accident. Estimates of the amount of plutonium released over the entire period from 10 p.m. to 2 a.m. are also discussed.

6. RESULTS

The results of calculations of plutonium release during 15-minute periods after the start of the fire are shown in Figure 6.1. For each time period, there is a distribution of estimates. The central (median, 50th percentile) value of each distribution is indicated by the horizontal bar. The confidence interval (bounded by the 5th and 95th percentiles of the distribution) for a particular estimate is indicated by the length of the vertical line. The figure shows that releases are estimated to have been largest during the two periods between 10:15 and 10:45 p.m. Releases during 15-minute periods after that time were estimated to diminish substantially. By 2 a.m., water had been applied to the plenum filter fire for 105 minutes and estimated releases from burning of plenum filters are seen to be much lower. The results shown in Figure 6.1 are considered to be the most reliable estimates that were made.

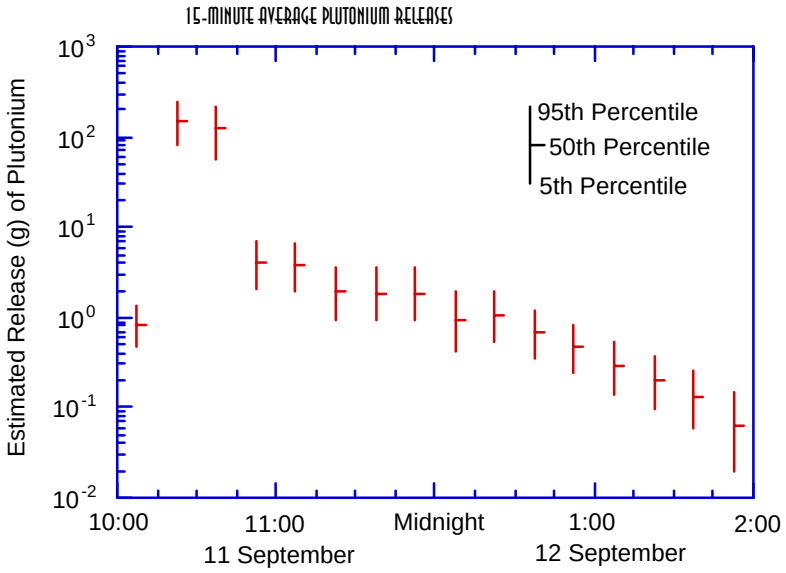

Figure 6.1. Estimated releases of plutonium to the atmosphere during consecutive 15-minute periods during the 1957 fire.

Although the chronology upon which the detailed estimates are based is not exact, it is useful to examine the more detailed estimates that were made for the first 45 minutes after the start of the fire. Those results are presented in Figure 6.2. It shows that the largest releases occurred during the periods when the main exhaust fans were operating at high speed and the filtration systems were seriously compromised. The changes in plenum filter efficiency are shown separately in Figure 5.2, and the period of rapid decline in plenum filter efficiency is also illustrated. The individual estimates shown in Figure 6.2 are subject to uncertainties in the times of the fire chronology. Totals for the longer (15-minute) periods are considered to be more reliable.

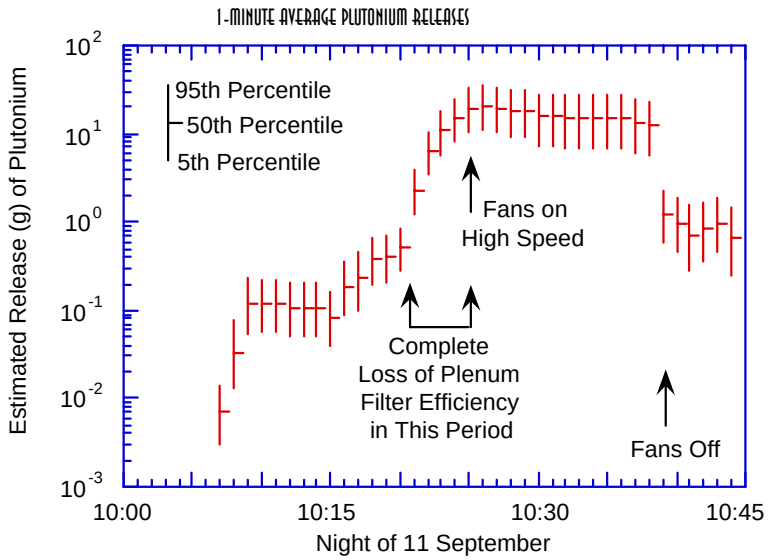

Figure 6.2. Estimated releases of plutonium to the atmosphere during consecutive 1-minute periods during the first 45 minutes of the 1957 fire.

The amount of plutonium that was released over the duration of the fire was estimated using the assumption of uniform distribution of metal into the four categories of metal oxidation. That assumption led to a median release estimate of ~300 g. The estimated confidence interval (5th to 95th percentiles) for that estimate was 160–490 g. This range does not reflect the scenario uncertainty identified in Section 5. The calculated releases are most sensitive to the amount of metal assumed to have been subject to very high temperature ("vigorous") oxidation. It is not known whether the necessary conditions were achieved during the fire. Additional calculations of the total release were performed using a range of assumptions regarding the amount of metal in the vigorous oxidation category. For those estimates, amounts of plutonium removed from that category were placed in the category of oxidation under dynamic conditions. These calculations indicate that the range of uncertainty in the total amount of plutonium released is from ~40 g to ~500 g when the scenario uncertainty is considered. In forming this range, we have rounded the upper bound given above (490 g) to one significant figure.

Table 6.1 shows a history of plutonium release estimates for the 1957 fire. The initial estimates were based upon the failure to detect appreciable amounts of plutonium in the environment during and soon after the fire. Considering the effective release height, many of the sampling locations chosen may well have been too close to the facility. The effective release height was affected both by the height of the stack and the rise of the hot plume because of its buoyancy and discharge velocity. The "large" estimates postulated in the pre-trial statement of the Church litigation (Fairfield and Woods 1978) and referenced by Johnson (1980) were based upon the mistaken belief that the results of Woodward (1971) were representative of conditions in 1953–1957 (see Section 3.2.3). The details of Barrick's calculations have not been located. They relied upon soil contamination measurements that were made several years after the 1957 fire.

Table 6.1. History of Estimates of Plutonium Releases from the 1957 Fire

Year	Source of release estimate	Release amount	Comments
1957	AEC[a]	"insignificant"	Plutonium was not mentioned in the public announcement. No release estimate was given in investigation report or in the serious accident bulletin.
1958–62	Dow internal reports	0.2–0.3 g	Amounts implied in descriptions of the history of plutonium releases from Building 71.
1970	Dow	"at least mCi amounts"	Answer to inquiry from AEC following publication of soil sampling report by Colorado Committee on Environmental Information.
1972	Dow Hammond presentation	"milligram amount" < 1 g offsite	Based upon environmental measurements.
1975–77	ERDA[b]/DOE	0.3–0.4 g	Amount of measured releases attributed to the fire; measurements did not include releases in the period 11–18 September.
1978	Plaintiffs' pre-trial statement[c]	"large amount"	Based upon their estimates of 10–250 kg of plutonium on plenum filters[d] and MUF of 14 kg.
1980	Johnson (1980)	"large"	No amount specified. Cited plaintiffs pre-trial statement estimates of the amount of plutonium on the main filter bank.
1981	Barrick (1981)	9.5–18 g	Based on interpolation and integration of soil contamination data (Krey and Hardy 1970).
1994	ChemRisk (1994b)	1 g (0.03–33 g)	Based upon environmental measurements.
1999	This report	40–500 g	Broad range resulting from uncertainty about conditions of plutonium oxidation (see text).

[a] U.S. Atomic Energy Commission.

[b] U.S. Energy Research and Development Administration; estimates prepared for Rocky Flats Environmental Impact Statement (DOE 1980).

[c] Documents from the Church lawsuit related to historic environmental contamination by releases from Rocky Flats facilities (Fairfield and Woods 1978).

[d] Estimates were based on filter contamination rates reported by Woodward (1971); see Section 3.2.3.

The size distributions of plutonium particles that were released at various times during the 1957 fire are not known. No measurements of the particle size of the airborne activity were performed. Experiments in which plutonium was oxidized and contaminated materials were burned have yielded a broad range of aerosol sizes. As noted in Section 4, particles with aerodynamic diameters that exceed 10 µm cannot penetrate into the pulmonary region of the lung. For the types of plutonium oxidation and combustion of contaminated materials that occurred during the 1957 fire, we recommend considering of a range of activity median aerodynamic diameters between 1 µm and 10 µm.

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